

Magnetic Susceptibility of Dirac Fermions, Bi-Sb Alloys, Interacting Bloch Fermions, Dilute Nonmagnetic Alloys, and Kondo Alloys

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Abstract

Wide ranging interest in Dirac Hamiltonian is due to the emergence of novel materials, namely, graphene, topological insulators and superconductors, the newly-discovered Weyl semimetals, and still actively-sought after Majorana fermions in real materials. We give a brief review of the relativistic Dirac quantum mechanics and its impact in the developments of modern physics. The quantum band dynamics of Dirac Hamiltonian is crucial in resolving the giant diamagnetism of bismuth and Bi-Sb alloys. Quantitative agreement of the theory with the experiments on Bi-Sb alloys has been achieved, and physically meaningful contributions to the diamagnetism has been identified. We also treat relativistic Dirac fermion as an interband dynamics in uniform magnetic fields. For the interacting Bloch electrons, the role of translation symmetry for calculating the magnetic susceptibility avoids any approximation to second order in the field. The magnetic susceptibility of Hubbard model and those of Fermi liquids are readily obtained as limiting cases. The expressions for magnetic susceptibility of dilute nonmagnetic alloys give a firm theoretical foundation of the empirical formulas used in fitting experimental results. For completeness, the magnetic susceptibility of dilute magnetic or Kondo alloys is also given for high and low temperature regimes.

Keywords: Dirac, Weyl, Majorana, Proca particles. BdG equation, topological superconductors. Foldy-Wouthuysen transformation, Magnetic susceptibility, interacting Bloch fermions, dilute alloys. Magnetic basis functions, Lattice Weyl transformation.

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I. INTRODUCTION

There is a growing interest from the nanoscience and nanotechnology community of the Dirac Hamiltonian in condensed matter owing to the emergence of novel materials that mimic the relativistic Dirac quantum mechanical behavior. The purpose of this review is to present

a unified treatment of Dirac Hamiltonian in solids and relativistic Dirac quantum mechanics from the point of view of energy-band quantum dynamics¹ coupled with the lattice Weyl transformation techniques.² This unified view seems to explicitly emerge in the calculation of the magnetic susceptibility of bismuth and Bi-Sb alloys.³ Large diamagnetism in solids has been attributed to interband quantum dynamics,⁴ often giving large g -factor due to pseudo-spin degrees of freedom and induced magnetic field. These are inherent in interband quantum dynamics. In most cases we are referring to two bands only which could be Kramer's degenerate bands.³⁻⁵ On the other hand, the classic Landau-Peierls diamagnetism is purely a single-band dynamical (orbital) effect. More recently, Fukuyama et al⁶ give a review on diamagnetism of Dirac electrons in solids from a theoretical perspective of many-body Green's function technique. However, no theoretical calculations were made and compared with the beautiful experiments of Wherli⁷ on the diamagnetism of Bi-Sb alloys.

Here we employ a theoretical perspective of band dynamics that has a long history even before the time of Peierls,⁴ who introduced the Peierls phase factor, and Wannier who introduced the Wannier function.⁸ This band dynamical treatment is generalized to the Dirac relativistic quantum mechanics and many-body condensed matter physics.⁹⁻¹² A detailed calculation¹³ of the diamagnetism of Bi-Sb alloys using the theory of Buot and McClure³ yields outstanding quantitative agreement with experimental results.⁷ We also give a review of the calculations of the magnetic susceptibility of other systems.

A. Historical Background

Firstly, in this section we will give a background on relativistic Dirac fermions, the beautiful Dirac equation and Dirac's declaration of anti-matter and discovery of positrons. We focus on its impact in motivating the development of modern physics, in particular condensed matter physics leading to a plethora of quasiparticle excitations with exotic properties. Because of this, condensed matter physics has become a low-energy proving ground on some of theoretical concepts in quantum field theory, high-energy elementary particle physics, and cosmology, where the Dirac equation has been extended and consistently deformed in ways exposing novel excitations/quasiparticles in physical systems.

In the space-time domain of condensed matter physics, it is interesting that the relativistic

Dirac-like equation was first recognized in the $\vec{k} \cdot \vec{p}$ band theory of bismuth and Bi-Sb alloys.¹⁴ This scientific historical event is like a repeat of what has happened in ordinary space-time with the quantum theory of relativistic electrons published in 1928 by Dirac¹⁵ in the form of what is now known as the Dirac equation with *four-component* fields. The following year, Weyl¹⁶ showed that for massless fermions, a simpler equation would suffice, involving *two-component* fields as opposed to the four-component fields of Dirac equation. These massless fermions is now known as the Weyl spin $\frac{1}{2}$ fermions. About nine years later, in 1937, Majorana¹⁷ was searching for a real version of the Dirac equation which is still Lorentz invariant. Thus, by imposing reality constraint of the Dirac equation, other solutions were obtained by Majorana still describing spin $\frac{1}{2}$ fermions, whose outstanding unique property is that they are their own anti-particles. By virtue of the fact that the complex field of the Dirac fermions is replaced by real fields, one refers to Dirac fermions as consisting of two Majorana fermions. Thus, Majorana fermions are often referred to as half-fermions.

Developments in physics in the early 20th century is not only confined to relativistic fermions but also to relativistic bosons which act as force-fields between particles. These particle-particle interactions are usually mediated by massless bosons such as photons, gluons, and gravitons. However, relativistic massive bosons, the so-called Proca particles, mediate the weak interactions between elementary particles. These are, for example, the W^\pm and Z^0 spin-1 heavy vector bosons. Twenty five years later after Majorana, Skyrme¹⁸ proposed a topological soliton in quantum field theory which is now referred to as skyrmion. Then in 1978, Callan et al¹⁹ proposed another topological objects known as merons. In magnetic systems, skyrmions, merons and bimerons are closely related.

Thus, whereas Weyl demonstrated the existence of massless relativistic spin- $\frac{1}{2}$ fermions, Proca²⁰ demonstrated in 1936 the existence of massive relativistic spin-1 bosons. In crystals, phonons are generally classified as the Nambu-Goldstone modes, but in the interaction between Cooper pairs in BCS superconductivity theory no massless phonons are present, only massive plasmalike excitations.²¹ In gauge-field theory of standard model, the Proca action is the gauge-fixed version of the Stueckelberg action which is a special case of Higgs mechanism through which the boson acquires mass.

Weyl fermions are irreducible representations of the proper Lorentz group, they are considered as building blocks of any kind of fermion field. Weyl fermions are either right chiral or left chiral but can not have both components. A general fermion field can be described

by two Weyl fields, one left-chiral and one right-chiral. It is worth mentioning that helicity and chirality coincide for massless fermions. By combining massless Weyl-fermion fields of different chiralities, one has not really generated a mass but has created a group-theoretical framework where mass can be allowed in the Dirac Lagrangian since the mass term must contain two different chiralities. Thus, a *massive* fermion must have a left-chiral as well as a right-chiral component.

1. *Parallel events in condensed matter physics*

Surprisingly the above chain of scientific events in ordinary space-time have been followed, although much later experimentally, by corresponding events in the space-time domain of condensed matter systems. Although, as early as 1937, Herring²² have already theoretically predicted the possibility of Weyl points in band theory of solid state physics. In more recent years Nielsen and Ninomiya²³ have suggested that excess of particles with a particular chirality were associated with Weyl fermions and could be observable in solids.

As mentioned before, the explicit form of Dirac Hamiltonian first appeared in bismuth and Bi-Sb alloys in a paper by Wolff in 1964 upon Blount's suggestion.¹⁴ Actually Cohen²⁴ gave the same form of the Hamiltonian of bismuth four years earlier in 1960, but did not cast into the form of Dirac Hamiltonian. Then, with the discovery of graphene²⁵ in 2004, massless Dirac fermions were identified in the energy-band structure at the K -points of the Brillouin zone. The following years are marked with understanding of materials with band structures that are “topologically” protected, typically of materials with strong spin-orbit coupling. This understanding have taken roots much earlier from works on quantum Hall effect (QHE) and quantum spin Hall effect (QSHE). The term ‘topological’ refers to a concept whereby there is a ‘holographic’ quasiparticle-state structure often localized at boundaries, domain walls or defects, which is topologically protected by the properties of the bulk. This maybe viewed as a sort of *entanglement* of the excitation-state with the bulk structure and therefore a *highly-nonlocal* property of excitation-state immune to local perturbation.²⁶

The pertinent measure of this entanglement is the subject of exploding research activities on the so-called *topological entanglement entropy*.²⁷ Indeed, a *holographic interpretation* of topological entanglement entropy was given.²⁸ This is also known in the literature simply as

generalized *bulk-boundary correspondence*. These years are replete with findings of topological insulators and topological superconductors, based on the realization that band theory must take into account concepts such as Chern numbers and Berry phases, familiar in quantum field theory of elementary particles, quantum-Hall effect, Peierls phase of a plaquette, Aharonov-Bohm effect, and in Born-Oppenheimer approximations. Indeed, theorists are now engaged in the exciting fields of topological field theory (TFT) and topological band theory (TBT). This attests to the merging of elementary particle physics, cosmology, and condensed matter physics.

Topological superconductors are simply the analogue of topological insulators. Whereas, topological insulators (TI) have a bulk band-gap with odd number of *relativistic Dirac fermions* and gapless modes on the surface, topological superconductors (TSC) are certain type of full superconducting gap TSC in the bulk, but due to the inherent particle-hole symmetry, have gapless modes of chargeless *Majorana ‘edge’ states*, also associated with *Andreev bound state* (ABS)^{29,30} on its boundaries, interfaces (e.g. interface between topological insulators and superconductors), and defects, supported by the bulk topological invariants. The emergence of topological insulators and superconductors also brought to light two types of quasiparticles properties, namely, (a) local or ‘trivial’ quasiparticles, and (b) nonlocal, ‘non-trivial’ or topological quasiparticles, often termed as *topological charge*. The second type are robust states, these cannot be created or removed by any local operators. The topological charge is also called the topological quantum number and is sometimes called the winding number of the solution. The topological quantum numbers are topological invariants associated with topological defects or soliton-type solutions of some set of differential equations modeling a physical system.

For example, the first 3D topological insulator was predicted for $\text{Bi}_{1-x}\text{Sb}_x$ system with ($0 \leq x \leq 0.04$), where the theoretical band structure calculation predicts the 3D topological insulator phase in $\text{Bi}_{0.9}\text{Sb}_{0.1}$.³¹ These developments are followed around mid-year of 2015 with the experimental discovery of Weyl fermions which were identified in the so-called Weyl semimetals. Specifically, the historic angle-resolved photoemission spectroscopy (ARPES) experiments performed on Ta-As has revealed Weyl fermions in the bulk.^{32,33} Likewise, similar experiments on photonic crystal have identified Weyl points (not Weyl fermions) inside the photonic crystal.³⁴ Weyl points differ from Dirac points since the former has two-component wavefunction whereas the later generally has four component wavefunction, e.g.

in graphene the K_{\pm} points in the Brillouin zone endows the two chiralities for a Dirac Hamiltonian for graphene.

The excitement about the experimental discovery of Weyl semimetals has to do with its great potential for ultrafast devices. The absence of backscattering for Weyl fermions is related to the so-called Klein paradox in quantum electrodynamics due to conservation of chirality. Because of this, Weyl fermions cannot be localized by random potential scattering in the form of Anderson localization³⁵ common to massive electrons. Moreover, with the efficient electron-hole pairs screening of impurities, mobilities of Weyl fermions are expected to be more than order of magnitude higher than the best Si transistors. In passing, we could say that the crossing of the bands of different symmetry properties in $\text{Bi}_{1-x}\text{Sb}_x$ alloys¹³ for antimony concentration of $x = 0.04$ might also serve as a Weyl semimetal, ignoring questions of topological stability.

a. The hunt for Majorana fermions On the other hand, the *experimental* Majorana fermions in solid state systems remains a challenging pursuit, a bit of ‘holy grail’ which one can perhaps draw a parallel with the search for Higgs particles in high-energy physics.³⁶ In conventional condensed matter system, c^{\dagger} and its Hermitian conjugate c is a *physically distinct operator* that annihilates electron or creates a hole. Since particles and antiparticles have opposite conserved charges, a Majorana fermion with its own antiparticle is a necessarily uncharged fermion. In his original paper, Majorana fermions can have arbitrary spin, so with spin zero it is still a fermion since the Majorana annihilation and creation operators still obey the anticommutation rule. For example, a mixture of particles and anti-particles of the form,

$$\gamma = \int dr \left(f(r)\Psi_{\uparrow} + g(r)\Psi_{\downarrow} + f^{*}(r)\Psi_{\downarrow}^{\dagger} + g^{*}(r)\Psi_{\uparrow}^{\dagger} \right) = \gamma^{\dagger}$$

indicates a chargeless and spinless Majorana fermions, often referred to as a *featureless* Majorana fermions. Another form of $\gamma = uc_{\sigma}^{\dagger} + u^{*}c_{\sigma}$ with equal spin projection, say a triplet or effectively spinless since spin degree of freedom does not have to be accounted for, is also a Majorana field operator, $\gamma = \gamma^{\dagger}$. This particular γ -form arises by imposing the Majorana condition³⁷ on the Bogoliubov-de Gennes (BdG) equation of superconductivity.

A conventional quasiparticle in superconductor is a broken Cooper pair, an excitation called a Bogoliubov quasiparticle and can have spin $\frac{1}{2}$. It is simply a linear combination of creation and annihilation operators, namely, $b_{\alpha} = u_{\alpha}a_{\alpha} + v_{\alpha}a_{-\alpha}^{\dagger}$ and $b_{-\alpha}^{\dagger} = u_{\alpha}^{*}a_{-\alpha}^{\dagger} + v_{\alpha}^{*}a_{\alpha}$,

where u_α and v_α are the components of the wavefunction of the Bogoliubov-de Gennes (BdG) equation. The requirement of Bogoliubov canonical transformation is that $[b_\alpha, b_{-\alpha}^\dagger] = (a_\alpha a_{-\alpha}^\dagger + a_{-\alpha}^\dagger a_\alpha)$, therefore we must have $(|u_\alpha|^2 + |v_\alpha|^2) = 1$ and $(u_\alpha v_\alpha^* + v_\alpha^* u_\alpha) = 0$. The particle created by the operator b_α^\dagger is a fermion, the so-called Bogoliubov quasiparticle (or ‘‘Bogoliubon’’). It combines the properties of a negatively charged electron and a positively charged hole. Indeed, Majorana fermion must have a form of superposition of particle and anti-particle. However, the creation and annihilation operators for Bogoliubons are still distinct. Thus, whereas charge prevents Majorana from emerging in a metal, on the other hand *distinct* creation and annihilation operators through superposition of electrons and holes with opposite spins is preventing Majorana quasiparticles in conventional *s*-wave superconductors. If Majorana fermion is to appear in the solid state it must therefore be in the form of *still to be experimentally demonstrated* nontrivial emergent Majorana excitations in real materials. The attention is focused on topological superconductors.

After a theoretical demonstration of the existence of Majoranas at the ends of a *p*-wave pairing Kitaev-chain, several theoretical demonstrations for the existence of zero-mode Majorana bound states (MBS) follow. In Kitaev’s prediction, inducing some types of superconductivity, known as the proximity effect, would cause the formation of Majoranas. These emergent particles are stable (Majorana degenerate bound states) and do not annihilate each other (unless the chain or wire is too short) because they are spatially separated. Thus, *p*-superconductors provide a natural hunting ground for Majoranas.

The search for Majorana has also paved the way for the *novel physics of zero modes* of the extended Dirac equation with inhomogeneous mass term that varies with position (corresponding to the momentum-dependent pairing potential in BdG equation), yielding a *kink-soliton* solution in 1-D, a *vortex* solution in 2-D, and a *magnetic monopole* in 3-D.³⁸ In condensed matter physics, the experimental search for Majorana is focused on exotic superconductors, namely, in triplet *p*-wave superconductivity in one dimension (1D), where MBS are located at both ends of the superconducting wire, and triplet *p + ip*-wave superconductivity in two dimensions (2D) where the MBS has been theoretically demonstrated to reside at the core of the vortex at an interface. In triplet 3D superconductors, the MBS is at the core of the ‘hedgehog’ configuration. These topological superconductors realize topological phases that support non-Abelian exotic excitations at their boundaries and at topological defects (e.g., hedgehog configuration). Most importantly, zero-energy modes localize at the

ends of a 1D topological p -wave superconductor, and bind to vortices in the 2D topological $p + ip$ -wave superconducting case. These zero-modes are precisely the condensed matter realization of Majorana fermions that are now being vigorously pursued. Moreover, engineered heterostructures using proximity effect with the s -wave superconductor, the so-called proximity-induced topological superconductor are correspondingly and vigorously also being pursued.

From the technological point of view these topologically-robust Majorana excitations are envisaged to implement quantum computing where braiding operation constitutes bits manipulation, analogous to the Yang-Baxter equations first introduced in statistical mechanics. The Majorana number density is limited to an integer (mod 2), i.e., 0 and 1, thus ideally representing a quantum bit. An intriguing proposal is a superconductor-topological insulator-superconductor (STIS) junction that forms a nonchiral 1D wire for Majorana fermions. These (STIS) junctions can be combined into circuits which allow for the creation, manipulation, and fusion of Majorana bound states for topological quantum computation.³⁹ There are also proposals for interacting non-Abelian anyons as Majorana fermions in Kitaev's honeycomb lattice model.⁴⁶ Indeed, Majorana fermions obey non-Abelian statistics, since Majorana fermions can have arbitrary spin statistics.

Several groups have experimentally reported detecting Majorana fermions.^{40–42} More recently, a Princeton group⁴³, have reported detecting Majorana by following Kitaev's prediction that, under the correct conditions a Majorana fermion bound states would appear at each end of a superconducting wire or Kitaev chain.

In summary, it is worth emphasizing that condensed matter physics has become the low-energy playground for discovering various quasiparticles and exotic topological excitations, which were mostly first proposed in quantum field theory of elementary particles, namely, Dirac fermions, Weyl fermions, Proca particles, vortices, skyrmions, merons, bimerons and other topologically-protected quasiparticles obeying non-Abelian and anyon statistical properties.

II. THE RELATIVISTIC DIRAC HAMILTONIAN

In ordinary space-time and in its original form, the Dirac equation is given by Dirac in the following forms⁴⁴

$$i\hbar \frac{\partial \psi(x, t)}{\partial t} = \left[\beta mc^2 + c \left(\sum_j^3 \alpha_j p_j \right) \right] \psi(x, t) \quad (1)$$

where

$$\beta = \begin{pmatrix} I_2 & 0 \\ 0 & -I_2 \end{pmatrix} = \gamma^0, \quad \alpha_\mu = \begin{pmatrix} 0 & \sigma_\mu \\ \sigma_\mu & 0 \end{pmatrix}, \quad \mu = 1, 2, 3$$

Therefore the Dirac Hamiltonian is of the matrix form is,

$$\mathcal{H}_{\text{Dirac}} = \begin{pmatrix} mc^2 & c\vec{p} \cdot \vec{\sigma} \\ c\vec{p} \cdot \vec{\sigma} & -mc^2 \end{pmatrix} \quad (2)$$

Equation (2) is the form that can occur in the $k \cdot p$ treatment of two-band theory of solids.

The Dirac Hamiltonian has eigenvalues given by

$$E = \pm \sqrt{m^2 c^4 + c^2 |p|^2}$$

Equation (1) can be rewritten as

$$i\hbar \beta \frac{\partial \psi(x, t)}{\partial ct} = \left[mc + \left(\sum_j^3 (\beta \alpha_j) p_j \right) \right] \psi(x, t)$$

and is usually given in its relativistic invariance form, as

$$i\hbar \gamma^\mu \partial_\mu \psi - mc\psi = 0 \quad (3)$$

where in the Dirac γ -basis,

$$\gamma^0 = \beta = \begin{pmatrix} I_2 & 0 \\ 0 & -I_2 \end{pmatrix}, \quad (4)$$

$$\gamma^\mu = \gamma^0 \alpha^\mu = \begin{pmatrix} 0 & \sigma_\mu \\ -\sigma_\mu & 0 \end{pmatrix} \quad (5)$$

These γ -matrices satisfy the relations of Clifford algebra,

$$\{\gamma^\mu, \gamma^\nu\} = 2\eta^{\mu\nu}$$

where the curly bracket stands for anticommutator. The anticommutator of $\{\sigma_x, \sigma_y\} = 2\delta_{xy}I$. Thus

$$\eta^{ij} = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & -1 & 0 & 0 \\ 0 & 0 & -1 & 0 \\ 0 & 0 & 0 & -1 \end{pmatrix} \quad (6)$$

defining Clifford algebra over a pseudo-orthogonal 4-D space with metric signature (1, 3) given by the matrix η^{ij} . It is a constant in special relativity but a function of space-time in general relativity. Equation (3) is an eigenvalue equation for the 4-momentum operator, $i\hbar\gamma^\mu\partial_\mu$ for the free Dirac electrons with eigenvalue equal to mc .

The Dirac γ -basis has the chirality operator given by,

$$\gamma^5 = \begin{pmatrix} 0 & I_2 \\ I_2 & 0 \end{pmatrix}$$

The number 5 is a remnant of old notation in which γ^0 was called “ γ^4 ”. Although γ^5 is not one of the gamma matrices of Clifford algebra over a pseudo-orthogonal 4-D space, this matrix is useful in discussions of quantum-mechanical chirality. For example, using the γ -matrices in the Dirac basis, a Dirac field can be projected onto its *left-handed* and *right-handed* components by,

$$\begin{aligned} \psi_L &= \frac{1}{2} (1 - \gamma^5) \psi \\ \psi_R &= \frac{1}{2} (1 + \gamma^5) \psi \end{aligned}$$

Thus, we have

$$\begin{aligned} \gamma^5 \psi_L &= -\psi_L \\ \gamma^5 \psi_R &= +\psi_R \end{aligned}$$

with eigenvalues ± 1 . The γ^5 anticommutes with all the γ^μ matrices.

On the other hand, the set $\{\gamma^0, \gamma^1, \gamma^2, \gamma^3, i\gamma^5\}$ forms the basis of the Clifford algebra in 5-spacetime dimensions for the metric signature (1, 4). The higher-dimensional γ -matrices generalize the 4-dimensional γ -matrices of Dirac to arbitrary dimensions. The higher-dimensional γ -matrices are utilized in *relativistically invariant* wave equations for fermions spinors in arbitrary space-time dimensions, notably in string theory and supergravity.

A. Weyl representation of Dirac equation

The Weyl representation of the γ -matrices is also known as the *chiral* basis, in which $\gamma^k (k = 1, 2, 3)$ remains the same but γ^0 is different, and so γ^5 is also different and diagonal. A possible choice of the Weyl basis is

$$\gamma^0 = \begin{pmatrix} 0 & -I_2 \\ -I_2 & 0 \end{pmatrix}, \quad \gamma^k = \begin{pmatrix} 0 & \sigma^k \\ -\sigma^k & 0 \end{pmatrix}, \quad \gamma^5 = \begin{pmatrix} I_2 & 0 \\ 0 & -I_2 \end{pmatrix}$$

In the *Weyl representation*, the Dirac equations reads

$$\begin{aligned} E\psi_1 &= c\vec{\sigma} \cdot \vec{p} \psi_1 + mc^2\psi_2 \\ E\psi_2 &= -c\vec{\sigma} \cdot \vec{p} \psi_2 + mc^2\psi_1 \end{aligned} \tag{7}$$

It is worthwhile to point out that Eq.(7) is interesting and bears resemblance to the eigenvalue equation for graphene if the \pm chirality degree of freedom of the zero-mode dispersions from the two inequivalent K_{\pm} points in the Brillouin zone (BZ) is taken into account. The isospin degree of freedom arises from the degeneracy of these inequivalent K_{\pm} points at the BZ corners. Thus, the K_{\pm} points Dirac electrons in graphene fits the Weyl representation of relativistic Dirac equations. It is the isospin degree of freedom that gives each K point in BZ a definite chirality. This has several exotic physical consequences as will be discussed in a separate paper by the authors.

Thus, in accounting for the K_{\pm} points of the Brillouin zone of graphene its Hamiltonian exactly resembles the relativistic Dirac Hamiltonian in the Weyl representation with zero mass. Note that if $m = 0$, we only need to solve one of the 2×2 matrix equations, yielding massless Weyl fermions with definite chirality (note also that chirality and helicity are both good quantum labels for massless fermions). This is clarified in what follows.

In matrix form we have,

$$E \begin{pmatrix} \psi_1 \\ \psi_2 \end{pmatrix} = c\vec{\sigma} \cdot p\gamma^5 \begin{pmatrix} \psi_1 \\ \psi_2 \end{pmatrix} - mc^2\gamma^0 \begin{pmatrix} \psi_1 \\ \psi_2 \end{pmatrix}$$

The eigenvalues are,

$$E = \pm \sqrt{c^2 p^2 + m^2 c^4}$$

Here, ψ_1 and ψ_2 are the eigenstates of the chirality operator γ^5 . The Weyl basis has the

advantage that its chiral projections take a simple form,

$$\begin{aligned}\psi_R &= \frac{1}{2}(1 - \gamma^5) \begin{pmatrix} \psi_1 \\ \psi_2 \end{pmatrix} = \begin{pmatrix} \psi_1 \\ 0 \end{pmatrix} \\ \psi_L &= \frac{1}{2}(1 + \gamma^5) \begin{pmatrix} \psi_1 \\ \psi_2 \end{pmatrix} = \begin{pmatrix} 0 \\ \psi_2 \end{pmatrix}\end{aligned}$$

Hence, in Weyl chirality γ -basis, we have

$$\gamma^5 \psi_R = \psi_R, \quad \gamma^5 \psi_L = -\psi_L$$

Thus chirality and helicity are a good quantum numbers for Weyl massless fermions.

B. Majorana representation of Dirac equation

The Majorana representation of Dirac equation can occur in p -wave superconductors. In the the Majorana γ -basis, all of the Dirac matrices are imaginary and spinors Ψ are real. We have

$$\hat{\gamma}^0 = \begin{pmatrix} 0 & \sigma_2 \\ \sigma_2 & 0 \end{pmatrix}, \quad (8)$$

$$\hat{\alpha}^1 = \gamma^0 \gamma^1 = - \begin{pmatrix} 0 & \sigma^1 \\ \sigma^1 & 0 \end{pmatrix}, \quad \hat{\gamma}^1 = \begin{pmatrix} i\sigma_3 & 0 \\ 0 & i\sigma_3 \end{pmatrix}, \quad (9)$$

$$\hat{\alpha}^2 = \gamma^0 \gamma^2 = \begin{pmatrix} I_2 & 0 \\ 0 & -I_2 \end{pmatrix}, \quad \hat{\gamma}^2 = \begin{pmatrix} 0 & -\sigma_2 \\ \sigma_2 & 0 \end{pmatrix}, \quad (10)$$

$$\hat{\alpha}^3 = \gamma^0 \gamma^3 = - \begin{pmatrix} 0 & \sigma^3 \\ \sigma^3 & 0 \end{pmatrix}, \quad \hat{\gamma}^3 = \begin{pmatrix} -i\sigma_1 & 0 \\ 0 & -i\sigma_1 \end{pmatrix}. \quad (11)$$

The gamma matrices are imaginary to obtain the particle-physics metric $(1, 3)$, i.e., $(+, -, -, -)$ in which squared masses are positive.

The Majorana relativistic equation is thus given by

$$i\hbar\gamma^\mu\partial_\mu\psi - mc\psi = 0$$

Using the relation $\alpha_\mu = \gamma^0\gamma^\mu$, we obtain after multiplying by γ^0 ,

$$i\hbar\gamma^0\gamma^\mu\partial_\mu\psi - \gamma^0mc\psi = 0$$

which reduces to

$$i\hbar \frac{\partial}{\partial t} \Psi = (c\hat{\alpha} \cdot p + \hat{\gamma}^0 mc^2) \Psi$$

where $p = -i\hbar\nabla$ is imaginary, $\hat{\alpha}$ is real, and $\hat{\gamma}^0 = \beta_{\text{majorana}}$ is imaginary. Thus, the Majorana relativistic equation is real, giving real solution Ψ , which ensures charge neutrality of spin $\frac{1}{2}$ particle which is its own antiparticle. Note that in Dirac equation the *Dirac mass* couples left- and right-handed chirality, whereas in Majorana equation, the *Majorana mass* couples particle with antiparticle.

In terms of matrix equation, we have

$$\begin{aligned} i\hbar \frac{\partial}{\partial t} \begin{pmatrix} \Psi_1 \\ \Psi_2 \end{pmatrix} &= (c\hat{\alpha}^\mu p_\mu + \hat{\gamma}^0 mc^2) \Psi \\ &= \begin{pmatrix} I_2 cp_y & -\sigma^1 cp_x - \sigma^3 cp_z + \sigma_2 mc^2 \\ -\sigma^1 cp_x - \sigma^3 cp_z + \sigma_2 mc^2 & -I_2 cp_y \end{pmatrix} \begin{pmatrix} \Psi_1 \\ \Psi_2 \end{pmatrix} \end{aligned} \quad (12)$$

Therefore we have coupled set of equations,

$$\begin{aligned} i\hbar \frac{\partial}{\partial t} \Psi_1 &= I_2 cp_y \Psi_1 - (\sigma^1 cp_x + \sigma^3 cp_z - \sigma_2 mc^2) \Psi_2 \\ i\hbar \frac{\partial}{\partial t} \Psi_2 &= -I_2 cp_y \Psi_2 - (\sigma^1 cp_x + \sigma^3 cp_z - \sigma_2 mc^2) \Psi_1 \end{aligned}$$

In $(2+1)$ -dimensional version, the matrix Hamiltonian of Eq.(12) can be written as

$$\mathcal{H}_M = \begin{pmatrix} I_2 cp_y & -\sigma^1 cp_x + \sigma_2 mc^2 \\ -\sigma^1 cp_x + \sigma_2 mc^2 & -I_2 cp_y \end{pmatrix}$$

By multiplying the wavefunction by a global phase equal to π , this can also be given by an equivalent expression,

$$\mathcal{H}_M = \begin{pmatrix} -I_2 cp_y & \sigma^1 cp_x - \sigma_2 mc^2 \\ \sigma^1 cp_x - \sigma_2 mc^2 & I_2 cp_y \end{pmatrix}.$$

In the case of Majorana fermions in superconductor, the Majorana mass term mc^2 corresponds to absolute value of the pair potential $|\Delta|$. However, in general $\Delta = \Delta_R + i\Delta_I$. Thus, to have a real Majorana equation in p -wave superconductor, we can expect the following form for the self-adjoint Majorana Hamiltonian in superconductor,⁴⁵

$$\mathcal{H}_M = \begin{pmatrix} -I_2 p_y & \sigma^1 p_x + iI_2 \Delta_I - \sigma_2 \Delta_R \\ \sigma^1 p_x - iI_2 \Delta_I - \sigma_2 \Delta_R & I_2 p_y \end{pmatrix}$$

where we have factored out the constant c or equated to unity. This can be substituted by a constant group velocity, v , for zero-gap or ‘massless’ states.

III. SUPERCONDUCTOR: BOGOLIUBOV-DE GENNES EQUATION

There is a formal analogy between the Dirac relativistic equation, BCS theory of superconductivity and BdG equation. We shall see later that their Hamiltonians have resemblance with the Hamiltonian of Bi and Bi-Sb alloys.

We can intuitively understand how the phonon-mediated electron-electron scattering in metals results in attractive interaction, i.e., by exchange of bosons leading to Cooper pairing. The instantaneous emission and absorption of highly-energetic phonons by interacting pair of electrons near the Fermi surface with opposite initial momentum, $-k$ and k , and with final momentum states of the Cooper pair in the form

$$-k + q \longleftrightarrow k - q$$

where q is the phonon wavevector will endow *opposite impulses* to the pair. On the average, this becomes an attractive-binding force between them, resulting in a zero-total-momentum BCS bound state. In general, this attractive interaction dominates in highly-dense-electron metal system with efficient Coulomb-potential screening. This condition yields nonzero mean-field average (‘pairing’) for $\langle \psi(x)\psi(x') \rangle$ and its complex conjugate $\langle \psi^\dagger(x)\psi^\dagger(x') \rangle$.

A. The BCS theory of superconductivity

In BCS theory, not only the momentum will have opposite sign but pairs must have opposite spin as well to maximize interaction, because the exchange interaction between parallel spins will reduce the attractive phonon-mediated interaction. Thus, the initial momentum of phonon-mediated interaction between pair of electron is of the set $\{\hbar\vec{k}_\uparrow, -\hbar\vec{k}_\downarrow\}$. This may also be interpreted as conservation of helicity for the pair. There are of course other boson-mediated pairing mechanisms which are more complex. For example, depending on the band structure a non-BCS pairing with nonzero total momentum of the pair in the form

$$-k + q \longleftrightarrow k + \kappa - q$$

where q is the phonon wavevector, or

$$-k + \kappa + q \longleftrightarrow k + \kappa - q$$

via spin-singlet channel are referred to as the Fulde-Ferrel-Larkin-Ovchinnikov (FFLO) pairing.^{49,50} The FFLO pairing was also proposed for *doped* Weyl semimetals which have a shifted Fermi surface brought by doping. The pairing theory was generalized to nonzero relative angular momentum type of pairing, such as the p -wave pairing, to be discussed later in connection with topological superconductors.

1. Effective BCS Hamiltonian

The simplest mean-field effective BCS many-body Hamiltonian can be rewritten in the equivalent *first-quantized* version through the BdG formalism. In the BdG formalism, the *eigenvalue problem* is essentially a first-quantized version of the second quantized effective BCS Hamiltonian formalism.

Consider a Hamiltonian of many-fermion system interacting through a spin-independent potential $\Phi(x)$,

$$H = \int \psi_{\sigma}^{\dagger}(x) H_0 \psi_{\sigma}(x) + \frac{1}{2} \sum_{\sigma\sigma'} \int \int d^3x d^3x' \left[\psi_{\sigma}^{\dagger}(x) \psi_{\sigma'}^{\dagger}(x') \Phi(x - x') \psi_{\sigma'}(x') \psi_{\sigma}(x) \right] \quad (13)$$

where σ is the spin index, $\Phi(x - x')$ is the translationally-invariant electron-phonon interaction potential, and

$$H_0 = -\frac{\hbar^2 \nabla^2}{2m} - \mu$$

since only the electrons near the Fermi surface can be redistributed or disturbed by the electron-electron interaction. Taking the Fourier transform to momentum space in finite volume V , we have

$$\psi_{\sigma}(x) = \frac{1}{\sqrt{V}} \sum_k a_k e^{ik \cdot x}$$

$$V(x) = \frac{1}{\sqrt{V}} \sum_q \Phi_q e^{iq \cdot x}$$

then we obtain

$$H = \sum_{k\sigma} \xi_k a_{k\sigma}^\dagger a_{k\sigma} + \frac{1}{2V} \sum_{\sigma\sigma'} \sum_{k,k',q} \Phi_q a_{k+q,\sigma}^\dagger a_{k'-q,\sigma'}^\dagger a_{k'\sigma'} a_{k\sigma}$$

where

$$\xi_k = E(k) - \mu$$

Restricting to pairing of fermions with zero total momentum and opposite spin, such that if k_\uparrow is occupied so is $-k_\downarrow$, we get the BCS Hamiltonian

$$H_{\text{BCS}} = \sum_{k\sigma} \xi_k a_{k\sigma}^\dagger a_{k\sigma} + \frac{1}{V} \sum_{k,k'} \Phi_{k-k'} a_{k',\uparrow}^\dagger a_{-k',\downarrow}^\dagger a_{-k,\downarrow} a_{k,\uparrow}$$

The bound pairs are not bose particles. We can define a creation and annihilation operators for pairs as follows

$$c_k = a_{-k,\downarrow} a_{k,\uparrow}, \quad c_k^\dagger = a_{k,\uparrow}^\dagger a_{-k,\downarrow}^\dagger, \quad c_k^\dagger c_k = n_{k,\uparrow} n_{-k,\downarrow}$$

we have

$$\begin{aligned} [c_k, c_{k'}^\dagger]_- &= (1 - n_{k\uparrow} - n_{-k\downarrow}) \delta_{kk'} \\ [c_k, c_{k'}]_- &= 0 \end{aligned}$$

where $n_{k\sigma} = a_{k,\sigma}^\dagger a_{k,\sigma}$, but the anticommutator given by

$$[c_k, c_{k'}]_+ = 2c_k c_{k'} (1 - \delta_{kk'})$$

is different from those of Bose particles. This is due to the terms, $(n_{k\uparrow} + n_{-k\downarrow})$ in $(1 - n_{k\uparrow} - n_{-k\downarrow})$ and $\delta_{kk'}$ in $(1 - \delta_{kk'})$ which comes from the Pauli exclusion principle. The Hamiltonian in terms of the c 's can be rewritten as

$$\begin{aligned} H_{\text{reduced}} &= \sum_{k\sigma} \xi_k c_{k\sigma}^\dagger c_{k\sigma} + \frac{1}{V} \sum_{k,k'} \Phi_{k-k'} c_{k'}^\dagger c_k \\ &= \sum_{k > k_F, \sigma} \epsilon_k c_{k\sigma}^\dagger c_{k\sigma} + \sum_{k < k_F, \sigma} |\epsilon_k| c_{k\sigma} c_{k\sigma}^\dagger \\ &\quad + \frac{1}{V} \sum_{k,k'} \Phi_{k-k'} c_{k'}^\dagger c_k - \sum_{k < k_F, \sigma} \epsilon_k (1 - n_{k\uparrow} - n_{-k\downarrow}) \end{aligned} \tag{14}$$

The effective BCS Hamiltonian in the mean-field approximation for c_k is obtained by writing

$$\Delta_k = -\frac{1}{V} \sum_{k'} \Phi_{k-k'} \langle c_{k'} \rangle$$

At finite temperature, the expression for the thermal average $\langle c_{k'} \rangle = \frac{\Delta_{k'}}{2E(k')} \tanh\left(\frac{\beta E_{k'}}{2}\right)$ yields the self-consistency condition for Δ_k , namely,

$$\Delta_k = -\frac{1}{V} \sum_{k'} \Phi_{k-k'} \frac{\Delta_{k'}}{2E(k')} \tanh\left(\frac{\beta E_{k'}}{2}\right) \quad (15)$$

Therefore Eq. (14) becomes in the mean field approximation,

$$\begin{aligned} H_{MF} &= \sum_{k\sigma} \xi_k a_{k\sigma}^\dagger a_{k\sigma} + \sum_k \Delta_k^* a_{-k,\downarrow} a_{k,\uparrow} + H.c. \\ &= \frac{1}{2} \sum_{k\sigma} \left(\xi_k a_{k\sigma}^\dagger a_{k\sigma} - \xi_k a_{-k\sigma} a_{-k\sigma}^\dagger \right) + \sum_k \Delta_k^* a_{-k,\downarrow} a_{k,\uparrow} + H.c. + \frac{1}{2} \sum_k \xi_k \end{aligned} \quad (16)$$

The spectrum of the last Hamiltonian can readily be found using the Nambu spinor,

$$A_k = \begin{pmatrix} a_{k,\uparrow} \\ a_{-k,\downarrow}^\dagger \end{pmatrix} \quad (17)$$

In terms of the Nambu spinor, the BCS Hamiltonian reads, by discarding irrelevant constant terms, as

$$\begin{aligned} H_{nambu} &= \sum_k A_k^\dagger \begin{pmatrix} \xi_k & \Delta_k \\ \Delta_k^* & -\xi_k \end{pmatrix} A_k \\ &= \frac{1}{2} \sum_{k\sigma} \left(\xi_k a_{k\sigma}^\dagger a_{k\sigma} - \xi_k a_{-k\sigma} a_{-k\sigma}^\dagger \right) + \sum_k \Delta_k^* a_{-k,\downarrow} a_{k,\uparrow} + H.c. \end{aligned}$$

The k -dependent spectrum, ϵ_k , can readily be calculated using the BdG first quantized equation, namely,

$$\begin{pmatrix} \xi_k & \Delta_k \\ \Delta_k^* & -\xi_k \end{pmatrix} \begin{pmatrix} u \\ v \end{pmatrix} = \epsilon_k \begin{pmatrix} u \\ v \end{pmatrix}$$

which yields

$$\epsilon_k = \pm \sqrt{\xi_k^2 + |\Delta_k|^2}$$

B. Bogoliubov Quasiparticles

We now expand A_k in terms of the eigenfunctions of the Hamiltonian. This is known as the *Bogoliubov transformation*. We have,

$$\hat{\Gamma} = U A_k$$

where U is the matrix of the eigenfunctions

$$U = \begin{pmatrix} u & -v \\ v & u \end{pmatrix}$$

where $|u|^2 + |v|^2 = 1$ by normality condition, and $u^*v - v^*u = 0$ by the orthogonality condition.

We therefore have

$$\begin{pmatrix} \gamma_{k,\uparrow} \\ \gamma_{-k,\downarrow} \end{pmatrix} = \begin{pmatrix} ua_{k,\uparrow} - va_{-k,\downarrow}^\dagger \\ ua_{-k,\downarrow}^\dagger + va_{k,\uparrow} \end{pmatrix} \\ \begin{pmatrix} \gamma_{k,\uparrow}^\dagger \\ \gamma_{-k,\downarrow}^\dagger \end{pmatrix} = \begin{pmatrix} u^*a_{k,\uparrow}^\dagger - v^*a_{-k,\downarrow} \\ u^*a_{-k,\downarrow} + v^*a_{k,\uparrow}^\dagger \end{pmatrix} \quad (18)$$

with inverse transformation as

$$\begin{pmatrix} a_{k,\uparrow} \\ a_{-k,\downarrow}^\dagger \end{pmatrix} = \begin{pmatrix} u\gamma_{k,\uparrow} + v\gamma_{-k,\downarrow} \\ u\gamma_{-k,\downarrow} - v\gamma_{k,\uparrow} \end{pmatrix}$$

Note that the Bogoliubov γ 's are explicitly combinations of *particle* and *antiparticle* operators, this is inherent in superconductivity physics. The commutation relation is

$$\begin{aligned} \{\gamma_{k,\uparrow}, \gamma_{k,\uparrow}^\dagger\} &= |u|^2 + |v|^2 = 1 \\ \{\gamma_{k,\uparrow}, \gamma_{-k,\downarrow}^\dagger\} &= 0 \end{aligned}$$

In terms of γ 's the BCS Hamiltonian can now be written as

$$H = \sum_k \left\{ \gamma_{k,\uparrow}^\dagger \xi_k \gamma_{k,\uparrow} - \gamma_{-k,\downarrow}^\dagger \xi_k \gamma_{-k,\downarrow} + \gamma_{k,\uparrow}^\dagger \Delta_k \gamma_{-k,\downarrow} + \gamma_{-k,\downarrow}^\dagger \Delta_k^* \gamma_{k,\uparrow} \right\} \quad (19)$$

which is the Hamiltonian for the Bogoliubov quasiparticles.

1. The Heisenberg equation of motion: first-quantized BdG equation

$$\begin{aligned} i\hbar \frac{\partial}{\partial t} \gamma_{k,\uparrow} &= [\gamma_{k,\uparrow}, H] \\ i\hbar \frac{\partial}{\partial t} \gamma_{-k,\downarrow} &= [\gamma_{-k,\downarrow}, H] \end{aligned} \quad (20)$$

We readily obtain the effective Schrödinger equation,

$$\begin{aligned} i\hbar \frac{\partial}{\partial t} \gamma_{k,\uparrow} &= [\gamma_{k,\uparrow}, H] \\ &= \xi_k \gamma_{k,\uparrow} + \Delta_k \gamma_{-k,\downarrow} \end{aligned} \quad (21)$$

$$\begin{aligned} i\hbar \frac{\partial}{\partial t} \gamma_{-k,\downarrow} &= [\gamma_{-k,\downarrow}, H] \\ &= (-\xi_k) \gamma_{-k,\downarrow} + \Delta_k^* \gamma_{k,\uparrow} \end{aligned} \quad (22)$$

Therefore in matrix form, we have the *first-quantized Schrödinger equation* known as the BdG equation,

$$i\hbar \frac{\partial}{\partial t} \begin{pmatrix} \gamma_{k,\uparrow} \\ \gamma_{-k,\downarrow} \end{pmatrix} = \begin{pmatrix} \xi_k & \Delta_k \\ \Delta_k^* & -\xi_k \end{pmatrix} \begin{pmatrix} \gamma_{k,\uparrow} \\ \gamma_{-k,\downarrow} \end{pmatrix} \quad (23)$$

with eigenvalues

$$\varepsilon_k = \pm \sqrt{\xi_k^2 + |\Delta_k|^2}$$

2. Eigenfunctions

We can determine the eigenfunctions by the BdG matrix equation, Eq. (23)

$$\varepsilon_k \begin{pmatrix} \gamma_{k,\uparrow} \\ \gamma_{-k,\downarrow} \end{pmatrix} = \begin{pmatrix} \xi_k & \Delta_k \\ \Delta_k^* & -\xi_k \end{pmatrix} \begin{pmatrix} \gamma_{k,\uparrow} \\ \gamma_{-k,\downarrow} \end{pmatrix}$$

which yields,

$$\frac{\gamma_{-k,\downarrow}}{\gamma_{k,\uparrow}} = -\frac{(\xi_k - \varepsilon_k)}{\Delta_k} = -\frac{(\xi_k - \pm \sqrt{\xi_k^2 + |\Delta_k|^2})}{\Delta_k} \quad (24)$$

$$\frac{\gamma_{k,\uparrow}}{\gamma_{-k,\downarrow}} = \frac{(\xi_k + \varepsilon_k)}{\Delta_k^*} = \frac{(\xi_k + \pm \sqrt{\xi_k^2 + |\Delta_k|^2})}{\Delta_k^*} \quad (25)$$

The above determines the component of the eigenfunction in terms of its ratio only.

3. Diagonalization by an orthogonal transformation

Consider the ‘geometric’ transformation U given by

$$U = \begin{pmatrix} \cos \theta & -\sin \theta \\ \sin \theta & \cos \theta \end{pmatrix}$$

We identify the following expressions:

$$(\xi_k - \epsilon_k) \cos \theta + \Delta \sin \theta = 0$$

which yields

$$\begin{aligned} \cos^2 \theta &= \frac{\Delta^2}{\Delta^2 + (\xi_k - \epsilon_k)^2} \\ \sin \theta \cos \theta &= \left(\frac{-(\xi_k - \epsilon_k) \Delta}{\Delta^2 + (\xi_k - \epsilon_k)^2} \right) \end{aligned}$$

and the alternate expressions

$$\Delta^* \cos \theta - (\xi_k + \epsilon_k) \sin \theta = 0$$

which yields,

$$\begin{aligned} \cos^2 \theta &= \left(\frac{(\xi_k + \epsilon_k)^2}{(\xi_k + \epsilon_k)^2 + \Delta^{*2}} \right) \\ \sin \theta \cos \theta &= \left(\frac{(\xi_k + \epsilon_k) \Delta^*}{(\xi_k + \epsilon_k)^2 + \Delta^{*2}} \right) \end{aligned}$$

Note that

$$1 + \frac{\Delta^{2*}}{(\xi_k + \epsilon_k)^2} = 1 + \frac{(\xi_k - \epsilon_k)^2}{\Delta^2}$$

so we have two equivalent expressions for $\cos^2 \theta$ and $\sin \theta \cos \theta$, which will be handy in the diagonalization that follows.

Having obtained the expression for the cosine and sine functions of θ , we now proceed to diagonalize the mean-field BdG Hamiltonian as

$$\begin{aligned} &\begin{pmatrix} \cos \theta & \sin \theta \\ -\sin \theta & \cos \theta \end{pmatrix} \begin{pmatrix} \xi_k & \Delta_k \\ \Delta_k^* & -\xi_k \end{pmatrix} \begin{pmatrix} \cos \theta & -\sin \theta \\ \sin \theta & \cos \theta \end{pmatrix} \\ &= \begin{pmatrix} \left\{ \begin{aligned} &[\xi_k \cos^2 \theta + \Delta \cos \theta \sin \theta] \\ &+ [\Delta_k^* \sin \theta \cos \theta - \xi_k \sin^2 \theta] \\ &- [\xi_k \sin \theta \cos \theta + \Delta_k \sin^2 \theta] \\ &+ [\Delta_k^* \cos^2 \theta - \xi_k \cos \theta \sin \theta] \end{aligned} \right\} & \left\{ \begin{aligned} &[-\xi_k \cos \theta \sin \theta + \Delta_k \cos^2 \theta] \\ &+ [-\Delta_k^* \sin^2 \theta - \xi_k \sin \theta \cos \theta] \\ &- [-\xi_k \sin^2 \theta + \Delta_k \sin \theta \cos \theta] \\ &+ [-\Delta_k^* \cos \theta \sin \theta - \xi_k \cos^2 \theta] \end{aligned} \right\} \end{pmatrix} \end{aligned}$$

We have for the diagonal elements

$$\left[\xi_k \cos^2 \theta + \Delta \cos \theta \sin \theta \right] + \left[\Delta_k^* \sin \theta \cos \theta - \xi_k \sin^2 \theta \right] = \left\{ \frac{(\xi_k + \epsilon_k)^2 + \Delta_k^{2*}}{(\xi_k + \epsilon_k)^2 + \Delta^{*2}} \right\} \epsilon_k = \epsilon_k \quad (26)$$

and

$$\begin{aligned}
& \left[\xi_k \sin^2 \theta - \Delta_k \sin \theta \cos \theta \right] - \left[\Delta_k^* \cos \theta \sin \theta + \xi_k \cos^2 \theta \right] \\
&= \left(\frac{1}{(\xi_k + \epsilon_k)^2 + \Delta_k^{*2}} \right) \left\{ - [\Delta_k^{*2} + (\xi_k + \epsilon_k)^2] \epsilon_k \right\} \epsilon_k \\
&= -\epsilon_k
\end{aligned} \tag{27}$$

One can also readily show that the off-diagonal elements are identically zero.

4. Chirality: Doubling the degrees of freedom

We can introduce chirality and helicity degrees of freedom⁵¹ for each energy band by extending the Nambu field operator, Eq. (17), to four components, namely,

$$\Psi_a \equiv \begin{pmatrix} a_{k,\uparrow} \\ a_{k,\downarrow} \\ a_{-k,\uparrow}^\dagger \\ a_{-k,\downarrow}^\dagger \end{pmatrix}$$

Thus, aside from the original particle-hole degrees of freedom we have now introduce the spin degrees of freedom. Consider simplifying the Hamiltonian as follows,

$$\begin{aligned}
H_a &= \Psi_a^\dagger \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & -1 & 0 \\ 0 & 0 & 0 & -1 \end{pmatrix} \Psi_a \\
H_a &= a_{k,\uparrow}^\dagger a_{k,\uparrow} + a_{k,\downarrow}^\dagger a_{k,\downarrow} - a_{-k,\uparrow}^\dagger a_{-k,\uparrow}^\dagger - a_{-k,\downarrow}^\dagger a_{-k,\downarrow}^\dagger
\end{aligned}$$

The BdG Hamiltonian for $|\Delta_k| = 0$ becomes

$$\begin{aligned}
H_{|\Delta_k|=0} &= \frac{1}{2} \begin{pmatrix} \xi_k & 0 & 0 & 0 \\ 0 & \xi_k & 0 & 0 \\ 0 & 0 & -\xi_{-k} & 0 \\ 0 & 0 & 0 & -\xi_{-k} \end{pmatrix} \\
&= \frac{1}{2} \xi_k \sigma_z \otimes I_2
\end{aligned}$$

5. The pairing potential

We write the pairing Hamiltonian as

$$\begin{aligned}
H_\Delta &= \Delta c_{k\uparrow}^\dagger c_{-k\downarrow}^\dagger + \Delta^* c_{-k\downarrow} c_{k\uparrow} \\
&= \frac{1}{2} \left[\Delta \left(c_{k\uparrow}^\dagger c_{-k\downarrow}^\dagger - c_{-k\downarrow}^\dagger c_{k\uparrow}^\dagger \right) + \Delta^* \left(c_{-k\downarrow} c_{k\uparrow} - c_{k\uparrow} c_{-k\downarrow} \right) \right]
\end{aligned}$$

In matrix notation, we have,

$$\begin{aligned}
H_\Delta &= \sum_k \Psi_a^\dagger \begin{pmatrix} 0 & 0 & 0 & \Delta \\ 0 & 0 & -\Delta & 0 \\ 0 & -\Delta^* & 0 & 0 \\ \Delta^* & 0 & 0 & 0 \end{pmatrix} \Psi_a \\
&= \sum_k \left\{ \Delta \left(a_{k,\uparrow}^\dagger a_{-k,\downarrow}^\dagger - a_{-k,\downarrow}^\dagger a_{k,\uparrow}^\dagger \right) + \Delta^* \left(a_{-k,\downarrow} a_{k,\uparrow} - a_{k,\uparrow} a_{-k,\downarrow} \right) \right\}
\end{aligned}$$

We have for complex Δ ,

$$\begin{aligned}
&\begin{pmatrix} 0 & 0 & 0 & \Delta \\ 0 & 0 & -\Delta & 0 \\ 0 & -\Delta^* & 0 & 0 \\ \Delta^* & 0 & 0 & 0 \end{pmatrix} \\
&= \begin{pmatrix} 0 & 0 & 0 & \Delta_R \\ 0 & 0 & -\Delta_R & 0 \\ 0 & -\Delta_R & 0 & 0 \\ \Delta_R & 0 & 0 & 0 \end{pmatrix} + \begin{pmatrix} 0 & 0 & 0 & i\Delta_I \\ 0 & 0 & -i\Delta_I & 0 \\ 0 & i\Delta_I & 0 & 0 \\ -i\Delta_I & 0 & 0 & 0 \end{pmatrix} \\
&= -\Delta_R \sigma_y \otimes \sigma_y - \Delta_I \sigma_x \otimes \sigma_y
\end{aligned}$$

where

$$\sigma_y \otimes \sigma_y = \begin{pmatrix} 0 & 0 & 0 & -1 \\ 0 & 0 & 1 & 0 \\ 0 & 1 & 0 & 0 \\ -1 & 0 & 0 & 0 \end{pmatrix}$$

$$\sigma_x \otimes \sigma_y = \begin{pmatrix} 0 & 0 & 0 & -i \\ 0 & 0 & i & 0 \\ 0 & -i & 0 & 0 \\ i & 0 & 0 & 0 \end{pmatrix}$$

Therefore

$$H_{BdG} = \xi_k \sigma'_z \otimes I_2 - \Delta_R \sigma'_y \otimes \sigma_y - \Delta_I \sigma'_x \otimes \sigma_y$$

where the prime pertains to the particle-hole degrees of freedom.

The Bogoliubov transformation, Eq. (18) can also be extended to account for the chirality and spin degrees of freedom. The extended BdG equation is

$$\begin{pmatrix} (\xi_k - \epsilon_k) & 0 & 0 & \Delta \\ 0 & (\xi_k - \epsilon_k) & -\Delta & 0 \\ 0 & -\Delta^* & -(\xi_k + \epsilon_k) & 0 \\ \Delta^* & 0 & 0 & -(\xi_k + \epsilon_k) \end{pmatrix} \begin{pmatrix} \gamma_{k,\uparrow}^\dagger \\ \gamma_{k,\downarrow}^\dagger \\ \gamma_{-k,\uparrow}^\dagger \\ \gamma_{-k,\downarrow}^\dagger \end{pmatrix} = 0$$

$$(\xi_k - \epsilon_k) \gamma_{k,\uparrow}^\dagger + \Delta \gamma_{-k,\downarrow}^\dagger = 0 \implies -\frac{(\xi_k - \epsilon_k)}{\Delta} = \frac{\gamma_{-k,\downarrow}^\dagger}{\gamma_{k,\uparrow}^\dagger}$$

$$(\xi_k - \epsilon_k) \gamma_{k,\downarrow}^\dagger - \Delta \gamma_{-k,\uparrow}^\dagger = 0 \implies \frac{(\xi_k - \epsilon_k)}{\Delta} = \frac{\gamma_{-k,\uparrow}^\dagger}{\gamma_{k,\downarrow}^\dagger}$$

$$-\Delta^* \gamma_{k,\downarrow}^\dagger - (\xi_k + \epsilon_k) \gamma_{-k,\uparrow}^\dagger = 0 \implies -\frac{(\xi_k + \epsilon_k)}{\Delta^*} = \frac{\gamma_{k,\downarrow}^\dagger}{\gamma_{-k,\uparrow}^\dagger}$$

$$\Delta^* \gamma_{k,\uparrow}^\dagger - (\xi_k + \epsilon_k) \gamma_{-k,\downarrow}^\dagger = 0 \implies \frac{(\xi_k + \epsilon_k)}{\Delta^*} = \frac{\gamma_{k,\uparrow}^\dagger}{\gamma_{-k,\downarrow}^\dagger}$$

C. $(p_x + ip_y)$ -Wave Pairing for Topological Superconductors

In the original BCS treatment, pairing of particles was in a relative s -wave state. However, the pairing theory was generalized to nonzero relative angular momentum type of pairing.

Indeed, p -wave pairing was observed in He^3 . The belief is that d -wave pairing occurs in heavy fermion and high- T_c superconductors.

It is for nonzero relative angular momentum pairing that the resulting BdG equations yield the form of the Majorana representation of Dirac equations.⁵² The effective BCS Hamiltonian in the mean-field approximation of the phonon-mediated interaction between electrons can thus be written in the form

$$H_F = \int \psi^\dagger(x) H_0 \psi(x) + \int \int \left[\Delta^*(x, x') \psi(x) \psi(x') + \Delta(x, x') \psi^\dagger(x) \psi^\dagger(x') \right] \quad (28)$$

The mean-field interaction via spin-singlet pairing, $\Delta(x, x')$, is

$$\Delta(x, x') = -g \langle \psi(x) \psi(x') \rangle$$

where g is a coupling constant. For scattering problems, it is often convenient to cast the Hamiltonian in momentum space. We have for the mean-field interaction,

$$\begin{aligned} \Delta_k &= -g \langle a_{-k\downarrow} a_{k\uparrow} \rangle && \text{for } s\text{-wave superconductivity} \\ &= \Delta(k_x - ik_y) && \text{for } (p_x + ip_y)\text{-wave superconductivity as } \vec{k} \Rightarrow 0 \end{aligned} \quad (29)$$

In the RHS of Eq. (29), Δ is a constant. Thus, BCS superconductors can be classified by the symmetry of the anomalous mean-field average in the boson-mediated electron-electron interaction $\Delta(x, x')$. For a 2-D $(p_x + ip_y)$ -wave topological triplet superconductors, we have,

$$H_{eff} = \int d^2k \left[(\varepsilon_k - \mu) c_k^\dagger c_k + \frac{1}{2} (\Delta_k^* c_{-k} c_k + \Delta_k c_k^* c_{-k}^*) \right]$$

where $\varepsilon_k \simeq \frac{k^2}{2m^*}$ is the quasiparticle kinetic energy and μ is the effective chemical potential. Δ_k is the gap function which is proportional to order parameter of the superconducting state. Again, we have the constraint provided by the Pauli exclusion principle, namely, only the electrons near the Fermi surface can be redistributed or disturbed by the electron-electron interaction. This in contrast, for example, for the case of excitons in semiconductors which involve the hydrogen-like pairing of holes on top of valence band and electrons at the bottom of the conduction band, although interband electron-electron pairing is quite possible in graphene⁴⁷ with zero energy-gap so that the Fermi surface coincide with the bottom of the conduction band and the top of the valence band. Indeed exotic superconductivity for graphene has been predicted upon doping with carriers.⁴⁸

The effective Heisenberg equation of motion of the Hamiltonian, Eq. (28), is thus given by

$$\begin{aligned} i\hbar \frac{\partial}{\partial t} \psi(z) &= [\psi(z), H_F] \\ i\hbar \frac{\partial}{\partial t} \psi^\dagger(z) &= [\psi^\dagger(z), H_F] \end{aligned}$$

We obtain,

$$i\hbar \frac{\partial}{\partial t} \psi(z) = (\varepsilon_k - \mu) \psi(z) + \int \Delta^*(z, x') \psi^\dagger(x') + \int \psi^\dagger(x) \Delta(x, z) \quad (30)$$

$$i\hbar \frac{\partial}{\partial t} \psi^\dagger(z) = \psi^\dagger(z) (\varepsilon_k - \mu) - \left\{ \int \Delta^*(z, x') \psi(x') + \int \psi(x) \Delta^*(x, z) \right\} \quad (31)$$

The above set of coupled equations, Eqs. (30)-(31) is still operator equations. By averting to first quantization, the above equations represent the BdG equations. This can be transformed to momentum space, e.g., using

$$\begin{aligned} \psi(z) &= \frac{1}{\sqrt{V}} \sum_k e^{ik \cdot z} a_k, \\ \psi^\dagger(z) &= \frac{1}{\sqrt{V}} \sum_k e^{-ik \cdot z} a_k^\dagger. \end{aligned}$$

For the more complex p -wave pairing, solving for the quasiparticle spectrum may require the use of Bethe *ansatz*.⁵²

IV. THE DIRAC HAMILTONIAN IN BISMUTH

In the space-time domain of condensed matter populated by Bloch electrons whose band dynamics is characterize by Wannier functions and Bloch functions, a Dirac-like Hamiltonian appeared in a paper published in 1964 by Wolff.¹⁴ In the presence of spin-orbit coupling, time reversal, and space inversion symmetry, the energy bands are doubly degenerate, known as Kramer's conjugates.

We are interested in the L -point of the Brillouin zone where the direct gap is small. Wolff give the Hamiltonian of bismuth, including spin-orbit coupling, in a Dirac form as,

$$\mathcal{H} = \beta \left(\frac{E_g}{2} \right) + \frac{(\Delta k)^2}{2m} I + \begin{pmatrix} 0 & \mathcal{H}_1 \\ \mathcal{H}_1 & 0 \end{pmatrix} \quad (32)$$

where

$$\mathcal{H}_1 = i\Delta k \sum_{\lambda=1}^3 K_{\lambda} \sigma_{\lambda}$$

where K_{λ} 's are determined by the matrix elements, including spin-orbit effects, of the velocity operator given by

$$\vec{\pi} = \frac{\vec{p}}{m} + \frac{\mu_o}{4mc} \left(\vec{s} \times \vec{\nabla} V \right),$$

where μ_o is the Bohr magneton. This gives the spin-orbit interaction correctly to order $\frac{e^2 \hbar^2}{64m^4 c^4}$ in the Hamiltonian, and is gauge invariant. Expanding \mathcal{H}_1 Eq. (32), we have for \mathcal{H}_1 given by Wolff,

$$\begin{aligned} \mathcal{H}_1 &= \Delta k \begin{pmatrix} \text{Re}(t) + i \text{Im}(t) & \text{Re}(u) + i \text{Im}(u) \\ -[\text{Re}(u) - i \text{Im}(u)] & \text{Re}(t) - i \text{Im}(t) \end{pmatrix} \\ &= \Delta k \begin{pmatrix} t & u \\ -u^* & t^* \end{pmatrix}. \end{aligned}$$

The total Hamiltonian is of the form

$$\mathcal{H} = \begin{pmatrix} \left(\frac{E_g}{2} \right) + \frac{(\Delta k)^2}{2m} & 0 & t & u \\ 0 & \left(\frac{E_g}{2} \right) + \frac{(\Delta k)^2}{2m} & -u^* & t^* \\ t & u & \left(-\frac{E_g}{2} \right) + \frac{(\Delta k)^2}{2m} & 0 \\ -u^* & t^* & 0 & \left(-\frac{E_g}{2} \right) + \frac{(\Delta k)^2}{2m} \end{pmatrix}$$

Wolff eliminated the $\text{Re}(t)$ by some unitary transformation applied to \mathcal{H}_1 . Upon substituting the matrix elements in terms of the basis $u_n(L, \Delta k = 0)$, we end up with the expression, where we explicitly indicates the symmetry types of the corresponding band-edge wavefunctions at the L -point of the Brillouin zone as,

$$\mathcal{H} = \begin{pmatrix} u_n(L, \Delta k = 0) & L_5 & L_6 & L_7 & L_8 \\ L_5 & \left(\frac{E_g}{2} \right) + \frac{(\Delta k)^2}{2m} & 0 & \Delta k \cdot \langle L_5 | \vec{\pi} | L_7 \rangle & \Delta k \cdot \langle L_5 | \vec{\pi} | L_8 \rangle \\ L_6 & 0 & \left(\frac{E_g}{2} \right) + \frac{(\Delta k)^2}{2m} & \Delta k \cdot \langle L_6 | \vec{\pi} | L_7 \rangle & \Delta k \cdot \langle L_6 | \vec{\pi} | L_8 \rangle \\ L_7 & \Delta k \cdot \langle L_7 | \vec{\pi} | L_5 \rangle & \Delta k \cdot \langle L_7 | \vec{\pi} | L_6 \rangle & \left(-\frac{E_g}{2} \right) + \frac{(\Delta k)^2}{2m} & 0 \\ L_8 & \Delta k \cdot \langle L_8 | \vec{\pi} | L_5 \rangle & \Delta k \cdot \langle L_8 | \vec{\pi} | L_6 \rangle & 0 & \left(-\frac{E_g}{2} \right) + \frac{(\Delta k)^2}{2m} \end{pmatrix}$$

The first two columns (rows) are for degenerate bands $\{L_5, L_6\}$ and for the last two columns (rows), for the degenerate $\{L_7, L_8\}$ at the L -point of the Brillouin zone. Observe that if

$\left(\pm \frac{E_g}{2}\right) + \frac{(\Delta k)^2}{2m} \implies 0$, we obtain the Weyl Hamiltonian. This condition holds at low energies with vanishing band-gap. This demonstrates the relative high-probability of finding Weyl fermions in solid state systems compared to finding Majorana fermions, which calls for more exotic quasiparticles that is yet to be found in real materials.

Earlier, Cohen in 1960 wrote the bismuth Hamiltonian²⁴ as

$$\mathcal{H} = \begin{pmatrix} K_0 - E_g & 0 & t & u \\ 0 & K_0 - E_g & -u^* & t^* \\ t^* & -u & K_1 & 0 \\ u^* & t & 0 & K_1 \end{pmatrix}$$

where the zero of energy is at the minimum of the conduction band.

For convenience in what follows, we recast the full $\vec{k} \cdot \vec{p}$ Hamiltonian as

$$\mathcal{H} = \begin{pmatrix} u_n(L) & L_5 & L_6 & L_7 & L_8 \\ L_5 & K_1 + \Delta & 0 & t & u^* \\ L_6 & 0 & K_1 + \Delta & -u & t^* \\ L_7 & t^* & -u^* & K_0 - \Delta & 0 \\ L_8 & u & t & 0 & K_0 - \Delta \end{pmatrix}$$

where the symmetry types of the corresponding band-edge wavefunctions of the first two columns (rows) are for degenerate bands $\{L_5, L_6\}$ and for the last two columns (rows), for the degenerate $\{L_7, L_8\}$ at the L -point of the Brillouin zone. Energies are measured from the center of the band gap, $\Delta = \frac{1}{2}E_g$, where E_g is the direct band gap at L -point, $K_1 = \frac{1}{2}k^2 + R_1$, $K_0 = \frac{1}{2}k^2 + R_0$ where R_1 and R_0 are contributions quadratic in k coming from bands other than the valence and conduction bands at L -point. The terms t and u are $\vec{k} \cdot \vec{\pi}$ matrix elements where $\vec{\pi} = \frac{\vec{p}}{m} + \frac{1}{2(mc)^2}(\vec{s} \times \vec{\nabla} V)$ includes the effect of spin-orbit coupling. The phases of t and u can be chosen independently without changing the form of the $\vec{k} \cdot \vec{p}$ Hamiltonian matrix. This fact allows for transformation of the above Hamiltonian to the Dirac form given by Wolff. We have

$$\begin{aligned} u &= \langle L_8 | \pi_y | L_5 \rangle k_y + \langle L_8 | \pi_z | L_5 \rangle k_z \\ &\equiv q_2 k_y + q_3 k_z \\ t &= \langle L_8 | \pi_x | L_6 \rangle k_x \end{aligned}$$

The coordinate axes are the binary (along Σ symmetry line), bisetrix (on the σ plane) and trigonal (along Λ symmetry line) crystal direction ($b, b, t, \implies x, y, z$ coordinate system). The eigenvalues of the $\vec{k} \cdot \vec{p}$ Hamiltonian matrix in the Lax two-band model, which neglect both K_1 and K_0 are

$$E = \pm \sqrt{\Delta^2 + |t|^2 + |u|^2}$$

where the $+$ and $-$ energy levels are doubly degenerate or Kramer conjugates. The reason for the neglect of K_1 and K_0 is that the most significant contribution to χ_\perp is χ_L^{22} when the magnetic field is along the bisetrix direction. Due to the large curvature of the energy bands in the binary and trigonal directions, the contribution of K_1 and K_0 are negligibly small. The Fermi surface is ellipsoidal and is tilted about the binary axis, there being a cross term in $k_y k_z$ from $|u|^2$. In the principal axes of the Fermi surface ellipsoid, the relation $\text{Re}(q'_2 q'_3) = 0$ holds. We therefore choose, $q'_1 = Q_1$, $q'_2 = -iQ_2$, and $q'_3 = Q_3$, where Q_1, Q_2 , and Q_3 are all real valued. We have in the principal axes,

$$\mathcal{H}' = \begin{pmatrix} u_n(L) & L'_5 & L'_6 & L'_7 & L'_8 \\ L'_5 & \Delta & 0 & Q_1 k_x & Q_3 k_z + iQ_2 k_y \\ L'_6 & 0 & \Delta & -Q_3 k_z + iQ_2 k_y & Q_1 k_x \\ L'_7 & Q_1 k_x & -Q_3 k_z - iQ_2 k_y & -\Delta & 0 \\ L'_8 & Q_3 k_z - iQ_2 k_y & Q_1 k_x & 0 & -\Delta \end{pmatrix} \quad (33)$$

We observe that rearranging the basis L'_7 and L'_8 we obtain

$$\mathcal{H}' = \begin{pmatrix} u_n(L) & L'_5 & L'_6 & L'_8 & L'_7 \\ L'_5 & \Delta & 0 & Q_3 k_z + iQ_2 k_y & Q_1 k_x \\ L'_6 & 0 & \Delta & Q_1 k_x & -Q_3 k_z + iQ_2 k_y \\ L'_8 & Q_3 k_z - iQ_2 k_y & Q_1 k_x & -\Delta & 0 \\ L'_7 & Q_1 k_x & -Q_3 k_z - iQ_2 k_y & 0 & -\Delta \end{pmatrix} \quad (34)$$

which has the form of the Bogoliubov-de Gennes (BdG) Hamiltonian of a 3-D topological superconductor which supports surface states as Majorana fermions.⁵³ The physics we are concern here is of course entirely different since we do not deal with boson-mediated electron-electron Cooper pairing.

A. Reduction of 4×4 Matrix to Diagonal Blocks of 2×2 Matrices

Rewrite Eq. (33) as

$$\begin{aligned}\mathcal{H}' &= \begin{pmatrix} u_n(L) & L'_5 & L'_6 & L'_7 & L'_8 \\ L'_5 & \Delta & 0 & \eta & \rho \\ L'_6 & 0 & \Delta & -\rho^* & \eta \\ L'_7 & \eta & -\rho & -\Delta & 0 \\ L'_8 & \rho^* & \eta & 0 & -\Delta \end{pmatrix} \\ &= \begin{pmatrix} D & A \\ A^{-1} & -D \end{pmatrix}\end{aligned}$$

The transformation \mathcal{U} is given by

$$\mathcal{U} = \begin{pmatrix} aI & -bI \\ bI & aI \end{pmatrix}$$

where unitarity condition renders

$$a^2 + b^2 = I$$

The other condition that determines a and b is the requirement that the transformed A' have zeros in the diagonal. Then one obtains

$$\mathcal{U}^{-1}\mathcal{H}'\mathcal{U} = \begin{pmatrix} u_n(L) & L''_5 & L''_6 & L''_7 & L''_8 \\ L''_5 & \varepsilon & 0 & 0 & \rho \\ L''_6 & 0 & \varepsilon & -\rho^* & 0 \\ L''_7 & 0 & -\rho & -\varepsilon & 0 \\ L''_8 & \rho^* & 0 & 0 & -\varepsilon \end{pmatrix}$$

We can rearrange the labels as

$$\tilde{\mathcal{H}} = \begin{pmatrix} u_n(L) & L''_5 & L''_8 & L''_6 & L''_7 \\ L''_5 & \varepsilon & \rho & 0 & 0 \\ L''_8 & \rho^* & -\varepsilon & 0 & 0 \\ L''_6 & 0 & 0 & \varepsilon & \rho^* \\ L''_7 & 0 & 0 & \rho & -\varepsilon \end{pmatrix} \quad (35)$$

where we have changed the sign of L_7'' , thus we obtain

$$\left(\tilde{\mathcal{H}}'\right) = \begin{pmatrix} H_1 & 0 \\ 0 & H_1^\dagger \end{pmatrix} \quad (36)$$

Therefore, we consider only the 2×2 Hamiltonian matrix H_1 in deriving the expression of the magnetic susceptibility. The $k \cdot p$ periodic eigenfunctions near the L -point of the 2×2 matrix H_1 are

$$\begin{aligned} L_c(k) &= aL_c + b^*L_v \\ L_v(k) &= aL_v - bL_c \end{aligned}$$

where

$$\begin{aligned} a &= \frac{\sqrt{E(E + \varepsilon)}}{\sqrt{2}E} \\ b &= \frac{Q_1k_x + iQ_3k_z}{\sqrt{2E(E + \varepsilon)}} \end{aligned}$$

The expression for the eigenvalues are

$$E_{\pm} = \pm \sqrt{\varepsilon^2 + |\rho|^2}$$

B. Magnetic Susceptibility of Dirac-Bloch Fermions

For calculating the magnetic susceptibility of Bloch electrons in bismuth with strong spin-orbit coupling, it is important that this should be accounted for in all stages of the calculations. This is described fully by the formalism given by Roth.⁵⁴ The susceptibility expression given by Roth can be written as a group of terms proportional to the first and second powers of Dirac-spin Bohr magneton plus remaining expression similar to that of Wannier and Upadhyaya⁵⁵ with $\vec{p} + \vec{k}$ replaced by $\vec{\pi} = \left(\vec{p} + \vec{k} + \frac{\beta}{c}\vec{s} \times \vec{\nabla}V\right)$ differing only in taking of traces due to the spin states in the wave function, but this is taken care of in our calculation by including H_1^\dagger also. The interaction of spin with the magnetic field in Roth's expression can be neglected since in the y -direction (small cyclotron mass direction) results in χ_L^{22} give the dominant contribution to χ_{\perp} , the susceptibility with the magnetic field *perpendicular* to the trigonal axis. This is also the direction where the simplified Lax two-band model is good for motion perpendicular to the magnetic field. One can neglect

$\vec{\mu}_0 \cdot \vec{B}$ in the effective Hamiltonian in this direction since the experimental g -factor due to *pseudo-spin* being of the order of 100 times that of free-electron spin moment. When the magnetic field is parallel to the trigonal axis, we denote the susceptibility as χ_{\parallel} .

C. Diamagnetism of Bismuth

After consolidating various terms in the susceptibility expression similar to the one given by Wannier and Upadhyaya,⁵⁵ Buot and McClure³ obtained a remarkably very simple expression for the most dominant contribution to $\chi_{\perp} \simeq \chi_L^{22}$ given by the expression

$$\chi_L^{22} = (6\pi^2 c^2)^{-1} \left(\frac{Q_3 Q_1}{Q_2} \right) \int_0^Z d\eta \frac{f(\varepsilon) - f(-\varepsilon)}{\varepsilon}$$

where

$$\begin{aligned} \eta &= Q_2 k_y \\ \varepsilon &= \sqrt{\eta^2 + \Delta^2} \end{aligned}$$

We can write

$$\begin{aligned} \chi_L^{22} &= (6\pi^2 c^2)^{-1} \left(\frac{Q_3 Q_1}{Q_2} \right) \int_0^Z d\eta \left[\frac{-1}{\varepsilon} + \left(\frac{f(\varepsilon) + 1 - f(-\varepsilon)}{\varepsilon} \right) \right] \\ &= \chi_{L,G}^{22} + \chi_{L,C}^{22} \end{aligned}$$

where $\chi_{L,G}^{22}$ is the large diamagnetic background term, independent of Fermi level and temperature, $\chi_{L,C}^{22}$ is the carrier paramagnetism and depends on Fermi level and temperature. $\chi_{L,G}^{22}$ and $\chi_{L,C}^{22}$ both depend on the energy gap, E_g , at symmetry point L . The other diagonal components, χ_L^{11} and χ_L^{33} are obtain by permutation of the Q_i 's. These are less significant than χ_L^{22} .

When the Fermi level lies in the forbidden gap and the temperature is low enough, then χ_{CP}^{22} , χ_{LP}^{22} , and $\chi_{L,C}^{22}$ are all zero and thus

$$\chi_{ID}^{22} = \chi_{L,G}^{22}$$

When the Fermi level is near the band edge at low temperature, then we have the following

relations

$$\begin{aligned}\chi_{LP}^{22} + \chi_{CP}^{22} &= \chi_{LC}^{22} \\ \chi_{ID}^{22} &= \chi_{LG}^{22} \\ \chi_{LP}^{22} &= -\frac{1}{3}\chi_{CP}^{22}\end{aligned}$$

The value of χ_{CP} is equal to the Pauli paramagnetism using the effective g -factor due to pseudospin moments. Similar relations hold for the other two principal directions of the magnetic field by simple rearrangement of the Q_i 's.

D. Diamagnetism of Bi-Sb Alloys

Using the known energy band structure, band parameters, and matrix elements consistent with experimental data on bismuth and Bi-Sb thus *implicitly including the spin-orbit coupling* in the $\vec{k} \cdot \vec{\pi}$ matrix elements, an outstanding fit of the calculated results with the detailed experimental data of Wherli on χ_{\perp} is obtained by Buot.¹³ This is shown in Fig. 1.

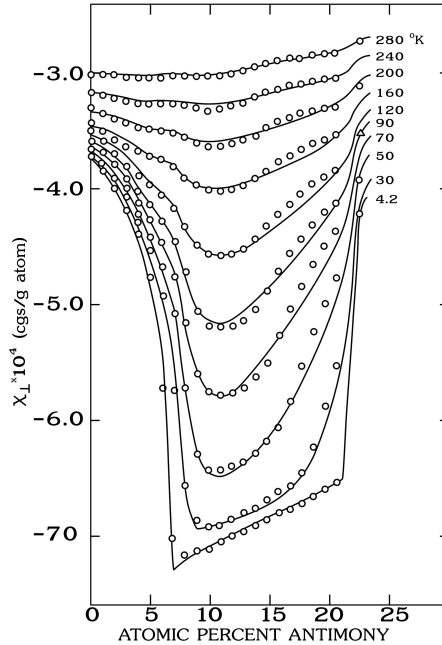


FIG. 1: Magnetic susceptibility, χ_{\perp} , at different temperatures perpendicular to the trigonal axis in $\text{Bi}_{1-x} - \text{Sb}_x$. The open circles are Wherli's experimental data for χ_{\perp} . The solid lines are the calculated data of Buot¹³ using the Buot and McClure theory.³ [Reproduced from Ref.¹³].

The susceptibility when the magnetic field is parallel to the trigonal axis, χ_{\parallel} , is also calculated. The main contribution comes from the T -point of the Brillouin zone. $\chi_{T,C}^{33}$ is calculated and $\chi_{T,G}^{33}$ adjusted to fit the experimental χ_{\parallel} data. $\chi_{T,G}^{33}$ is the contribution of the rest of the filled bands associated with symmetry point T over and above the contribution at point L . The calculated result for χ_{\parallel} compared with the experimental data of Wherli is shown in Fig. 2.

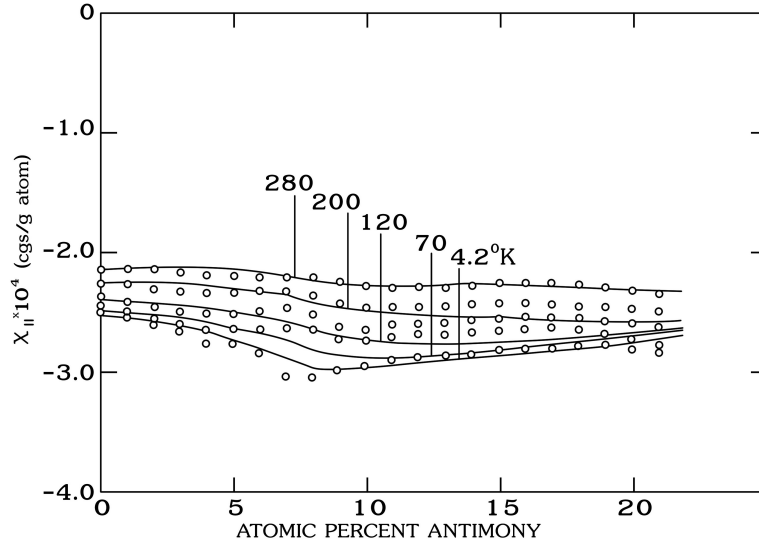


FIG. 2: Magnetic susceptibility parallel to the trigonal axis in $\text{Bi}_{1-x} - \text{Sb}_x$. The open circles are Wherli's experimental data for χ_{\parallel} . The solid lines represents the calculated data using the Buot and McClure theory. [Reproduced from Ref.¹³]

The large diamagnetism of bismuth is only incidentally related to the spin-orbit coupling since the band dynamical effects dominate. In fact the same form of the Hamiltonian as in Eqs. (35) and (36) applies at the H -point of graphite (without spin-orbit coupling) and also gives a large diamagnetism.^{3,56}

V. BAND THEORY OF MAGNETIC SUSCEPTIBILITY OF RELATIVISTIC DIRAC FERMIONS

In this section, we formulate the magnetic susceptibility of relativistic Dirac fermions analogous to energy-band dynamics of crystalline solids. The Hamiltonian of free relativistic

Dirac fermions is of the form

$$\mathcal{H} = \beta\Delta + c\vec{\alpha} \cdot \vec{P} \quad (37)$$

We designate quantum operators in capital letters and their corresponding eigenvalues in small letters. The equation for the eigenfunctions and eigenvalues is

$$\mathcal{H}b_\lambda(x, p) = E_\lambda(p)b_\lambda(x, p) \quad (38)$$

where $E_\lambda(p) = \pm E(p)$, and $E_\lambda(\vec{q}' - q) = \frac{1}{(2\pi\hbar)^3} \int d\vec{p} e^{(\frac{i}{\hbar})\vec{p} \cdot (\vec{q}' - q)} E_\lambda(p)$, λ labels the band index: \pm spin band for positive energy states and \pm spin band for negative energy states.

$$E_\lambda(p) = \pm \sqrt{(cp)^2 + (mc^2)^2}$$

The doubly degenerate bands is reminiscent of the Kramer conjugates in bismuth and Bi-Sb alloys. The localized function $a_\lambda(\vec{x} - \vec{q}')$ is the ‘Wannier function’ for relativistic Dirac fermions, defined below.⁵⁷

In the absence of magnetic field we may define the Wannier function and Bloch function of a relativistic Dirac fermions as

$$b_\lambda(x, p) = \frac{1}{(2\pi\hbar)^{\frac{3}{2}}} e^{(\frac{i}{\hbar})\vec{p} \cdot \vec{x}} u_\lambda(\vec{p})$$

$$a_\lambda(\vec{x} - \vec{q}) = \frac{1}{(2\pi\hbar)^{\frac{3}{2}}} \int d\vec{p} e^{(\frac{i}{\hbar})\vec{p} \cdot \vec{q}} b_\lambda(x, p)$$

where $b_\lambda(x, p)$ is the Bloch function, and $a_\lambda(\vec{x} - \vec{q})$ the corresponding Wannier function. $u_\lambda(\vec{p})$ is a four-component function. The $u_\lambda(\vec{p})$ ’s are related to the $u_\lambda(0)$ ’s by a unitary transformation, S , which also transforms the Dirac Hamiltonian into an *even* form, i.e., no longer have interband terms. This is equivalent to the transformation from Kohn-Luttinger basis to Bloch functions in $\vec{k} \cdot \vec{p}$ theory. We have

$$S = \frac{E + \beta\mathcal{H}}{\sqrt{2E(E + \Delta)}}$$

which can be written in matrix form as

$$S = \begin{pmatrix} \sqrt{\frac{(E+\Delta)}{2E}} & \frac{c\{\vec{\sigma} \cdot \vec{p}\}^*}{\sqrt{2E(E+\Delta)}} \\ -\frac{c\{\vec{\sigma} \cdot \vec{p}\}^*}{\sqrt{2E(E+\Delta)}} & \sqrt{\frac{(E+\Delta)}{2E}} \end{pmatrix}$$

where the entries are 2×2 matrices, $\Delta = mc^2$, and all matrix elements may be viewed as matrix elements of S between the $u_\lambda(0)$ ’s, which are the spin functions in the Pauli

representation. The transformed Hamiltonian is

$$\mathcal{H} = S\mathcal{H}S^\dagger = \beta E(\vec{P}) \quad (39)$$

The $a_\lambda(\vec{x} - \vec{q})$ is not a δ -function because of the dependence of $u_\lambda(\vec{p})$ on \vec{p} ; it is spread out over a region of the order of the Compton wavelength, $\frac{\hbar}{mc}$, of the electron and no smaller, as pointed out first by Newton and Wigner⁵⁹, Foldy and Wouthuijsen⁵⁸ and by Blount.⁸

The Weyl correspondence for the momentum and coordinate operator giving the correct dynamics of quasiparticles is given by the prescription that the momentum operator \vec{P} and coordinate operator \vec{Q} be defined with the aid of the Wannier function and the Bloch function as

$$\begin{aligned} \vec{P}b_\lambda(x, p) &= \vec{p}b_\lambda(x, p) \\ \vec{Q}a_\lambda(\vec{x} - \vec{q}) &= \vec{q}a_\lambda(\vec{x} - \vec{q}) \end{aligned}$$

and the uncertainty relation follows in the formalism,

$$[Q_i, P_j] = i\hbar\delta_{ij}$$

From Eq. (38), we have

$$\begin{aligned} \frac{1}{(2\pi\hbar)^{\frac{3}{2}}} \int dq e^{(-\frac{i}{\hbar})\vec{p}\cdot\vec{q}} \mathcal{H}a_\lambda(\vec{x} - \vec{q}) &= E_\lambda(p) \frac{1}{(2\pi\hbar)^{\frac{3}{2}}} \int dq e^{(-\frac{i}{\hbar})\vec{p}\cdot\vec{q}} a_\lambda(\vec{x} - \vec{q}) \\ \mathcal{H}a_\lambda(\vec{x} - \vec{q}') &= \int dq E_\lambda(\vec{q}' - q) a_\lambda(\vec{x} - \vec{q}) \end{aligned}$$

These relations allows us to transform the ‘bare’ Hamiltonian operator to an ‘effective Hamiltonian’ expressed in terms of the \vec{P} operator and the \vec{Q} operator. This is conveniently done by the use of the ‘lattice’ Weyl transform² (‘lattice’ Weyl transform and Weyl transform will be used interchangeably for infinite translationally invariant system including crystalline solids). Thus, any operator $A(\vec{P}, \vec{Q})$ which is a function of \vec{P} and \vec{Q} can be obtained from the matrix elements of the ‘bare’ operator, A_{op}^b , between the Wannier functions or between

the Bloch functions as,

$$\begin{aligned}
A(\vec{P}, \vec{Q}) &= \sum_{\lambda\lambda'} \int d\vec{v} d\vec{u} a_{\lambda\lambda'}(\vec{u}, \vec{v}) \exp \left[\left(-\frac{i}{\hbar} \right) \left(\vec{Q} \cdot \vec{u} + \vec{P} \cdot \vec{v} \right) \right] \Omega_{\lambda\lambda'} \\
a_{\lambda\lambda'}(\vec{u}, \vec{v}) &= h^{-8} \int d\vec{p} d\vec{q} a_{\lambda\lambda'}(\vec{p}, \vec{q}) \exp \left[\left(-\frac{i}{\hbar} \right) \left(\vec{q} \cdot \vec{u} + \vec{p} \cdot \vec{v} \right) \right] \\
a_{\lambda\lambda'}(\vec{p}, \vec{q}) &= \int d\vec{v} e^{\frac{i}{\hbar} \vec{p} \cdot \vec{v}} \left\langle \vec{q} - \frac{1}{2} \vec{v}, \lambda \left| A_{op}^b \right| \vec{q} + \frac{1}{2} \vec{v}, \lambda' \right\rangle \\
&= \int d\vec{u} e^{\frac{i}{\hbar} \vec{q} \cdot \vec{u}} \left\langle \vec{p} + \frac{1}{2} \vec{u}, \lambda \left| A_{op}^b \right| \vec{p} - \frac{1}{2} \vec{u}, \lambda' \right\rangle
\end{aligned}$$

where $|\vec{p}, \lambda\rangle$ and $|\vec{q}, \lambda\rangle$ are the state vectors representing the Bloch functions and Wannier functions, respectively, and

$$\begin{aligned}
\Omega_{\lambda\lambda'} &= \int d\vec{p} |\vec{p}, \lambda\rangle \langle \vec{p}, \lambda'| \\
&= \int d\vec{q} |\vec{q}, \lambda\rangle \langle \vec{q}, \lambda'|
\end{aligned}$$

A. Canonical Conjugate Dynamical Variables in Band Quantum Dynamics

A few more words about \vec{Q} and \vec{P} . The use of \vec{Q} , conjugate to the operator \vec{P} of the Hamiltonian in even form, is preferred in the band-dynamical formalism.¹ The reason we now associate \vec{Q} with the operator \vec{P} of the Hamiltonian in even form is that this momentum operator now belongs to the respective bands (each of infinite width) of the *decoupled* Dirac Hamiltonian. This operator is now analogous to the crystal momentum operator in crystalline solids. For the original Dirac Hamiltonian $\dot{x} = c$ [from Eq. (37)] leading to a complex *zitterbewegung* motion in x -space, whereas for the Hamiltonian in even form $\dot{Q} = v$ [from Eq. (39)], c is the speed of light and v the velocity of a wave packet in the classical limit, and thus Q is more closely related to the band dynamics of fermions than x . Moreover, on the cognizance that the continuum is the limit when the lattice constant of an array of lattice points goes to zero, there is a more compelling fundamental basis for using the lattice-position operator Q .⁵⁷ Since quantum mechanics is the mathematics of measurement processes,⁶⁰ the most probable measured values of the positions are the lattice-point coordinates. Indeed, these lattice points, or atomic sites, are where the electrons spend some time in crystalline solids. Therefore the *lattice points* and *crystal momentum* are clearly the *observables* of the theory and q and p constitute the eigenvalues of the lattice-point position

operator Q and crystal momentum operator P , respectively. Thus, Q is considered here as the generalized position operator in quantum theory for describing energy-band quantum dynamics, canonical conjugate to ‘crystal’ momentum operator \vec{P} of the Hamiltonian in even form. Although the ‘bare’ operator x can still be used as position operator it only unnecessarily renders very complicated and almost intractable resulting expressions,^{8,9} since this does not directly reflect the appropriate observables in band dynamics as first enunciated by Newton and Wigner⁵⁹ and by Wannier several decades ago.¹ Thus, in understanding the dynamics of Dirac relativistic quantum mechanics succinctly, position space should be defined at discrete points q which are eigenvalues of the operator Q .⁵⁷

B. The Even Form of Dirac Hamiltonian in a Uniform Magnetic Field

The Dirac Hamiltonian for an electron with anomalous magnetic moment in a magnetic field is

$$\mathcal{H}_{op} = \vec{\alpha} \cdot \vec{\Pi}_{op} + \beta mc^2 - \frac{1}{2}(g-2)\mu_B\beta\vec{\sigma} \cdot \vec{B}$$

where

$$\begin{aligned}\vec{\Pi}_{op} &= c\vec{P}_{op} - e\vec{A}(\vec{Q}_{op}) \\ \mu_B &= \frac{e\hbar}{2mc}\end{aligned}$$

The transformed Hamiltonian in even form \mathcal{H}'_B is given by Ericksen and Kolsrud¹⁰

$$\mathcal{H}'_B = \beta \left[m^2 c^4 + \Pi^2 - e\hbar c(1 + \lambda')\vec{\sigma} \cdot \vec{B} + \beta \left(\frac{\lambda' e\hbar}{2mc} \right) \sigma \cdot (B \times \Pi - \Pi \times B) \right]^{\frac{1}{2}} \quad (40)$$

where $\lambda' = \frac{1}{2}(g-2)$, and

$$\begin{aligned}\tilde{\Pi} &= cP - eA(Q) - eA(r) \\ &= cP - eA(Q+r)\end{aligned}$$

$$\begin{aligned}A(Q+r) &= \frac{1}{2}B \times (Q+r) \\ r &= \beta \left(\frac{\lambda' \hbar}{mc} \right) \sigma\end{aligned}$$

The above Hamiltonian can be written as

$$\begin{aligned}
\mathcal{H}'_B &= \beta \left[m^2 c^4 + \Pi^2 - e\hbar c(1 + \lambda') \vec{\sigma} \cdot \vec{B} - 2 \left(\frac{1}{2} B \times r \cdot \Pi \right) \right]^{\frac{1}{2}} \\
&= \beta \left[m^2 c^4 + \Pi^2 - e\hbar c(1 + \lambda') \vec{\sigma} \cdot \vec{B} - 2A(r) \cdot \Pi \right]^{\frac{1}{2}} \\
&= \beta \left[m^2 c^4 + \tilde{\Pi}^2 - e\hbar c(1 + \lambda') \vec{\sigma} \cdot \vec{B} - A^2(r) \right]^{\frac{1}{2}} \\
&= \beta \left[m^2 c^4 + \tilde{\Pi}^2 - e\hbar c(1 + \lambda') \vec{\sigma} \cdot \vec{B} - \left(\frac{\lambda' e \hbar}{2mc} \right)^2 B^2 \right]^{\frac{1}{2}}
\end{aligned} \tag{41}$$

C. Translation operator, $T_M(q)$, under uniform magnetic fields

In the presence of a uniform magnetic field, magnetic Wannier Functions, $A_\lambda(x - q)$, and magnetic Bloch functions, $B_\lambda(x, p)$, exist. This is proved by using symmetry arguments. In general, these two basis functions are complete and span all the eigensolutions of the magnetic Hamiltonian belonging to a band index λ . The magnetic Wannier Functions $A_\lambda(x - q)$ and magnetic Bloch functions $B_\lambda(x, p)$ are related by similar unitary transformation in the absence of magnetic field, namely,

$$\begin{aligned}
B_\lambda(x, p) &= \frac{1}{(2\pi\hbar)^{\frac{3}{2}}} e^{(\frac{i}{\hbar})\vec{p} \cdot \vec{x}} u_\lambda(\vec{p}) \\
A_{\lambda}(\vec{x} - \vec{q}) &= \frac{1}{(2\pi\hbar)^{\frac{3}{2}}} \int d\vec{p} e^{(\frac{i}{\hbar})\vec{p} \cdot \vec{q}} B_\lambda(x, p)
\end{aligned}$$

where \vec{p} and \vec{q} are quantum labels.

Under a uniform magnetic fields, we have for a translation operator, $T_M(q)$, obeying the relation,

$$\begin{aligned}
\nabla_r T_M(q) &= [P, T_M(q)] \\
&= \frac{ie}{\hbar c} A(q) T_M(q)
\end{aligned} \tag{42}$$

Therefore,

$$T_M(q) = \exp \left(\frac{-ie}{\hbar c} A(r) \cdot q \right) C(q)$$

where $C_0(q)$ is an operator which do not depend explicitly on r . Since $T_M(q)$ is a translation operator by amount q leads us to write

$$C_0(q) = \exp(-q \cdot \nabla_r), \quad \text{a pure displacement operator by amount } -q$$

Equation (42) means that $[P, T_M(q)]$ is diagonal if $T_M(q)$ is diagonal, and therefore they have the same eigenfunctions and the same quantum label. Therefore displacement operator in a translationally symmetric system under a uniform magnetic field acquire the so-called ‘*Peierls phase factor*’.

Clearly, bringing the wavepacket or Wannier function around a closed loop, or around plaquette in the tight-binding limit, would acquire a phase equal to the magnetic flux through the area defined by the loop. This is the so-called Bohm-Aharonov effect or Berry phase. Thus, the concept of Berry phase has actually been floating around in the theory of band dynamics since the time of Peierls. Berry⁶¹ has brilliantly generalized the concept to parameter-dependent Hamiltonians even in the absence of magnetic field through the so-called *Berry connection*, *Berry curvature*, and *Berry flux*.

The magnetic translation operator generates all magnetic Wannier functions belonging to band index λ from a given magnetic Wannier function centered at the origin, $A_\lambda^0(r-0)$, as

$$\begin{aligned} A_\lambda(r-q) &= T_M(q)A_\lambda^0(r-0) \\ &= \exp\left(\frac{-ie}{\hbar c}A(r)\cdot q\right)A_\lambda^0(r-q) \end{aligned}$$

We also have the following relation,

$$\begin{aligned} T_M(q)T_M(\rho) &= \exp\left(\frac{ie}{\hbar c}A(q)\cdot\rho\right)T_M(q+\rho) \\ [T_M(q), T_M(\rho)] &= \exp\left(\frac{ie}{\hbar c}A(q)\cdot\rho\right)T_M(q+\rho) - \exp\left(\frac{ie}{\hbar c}A(\rho)\cdot q\right)T_M(\rho+q) \\ &= 2i \sin\left(\frac{e}{\hbar c}A(q)\cdot\rho\right)T_M(q+\rho) \end{aligned}$$

Moreover, we have,

$$\begin{aligned} \mathcal{H}B_\lambda(x, p) &= E_\lambda\left(p - \frac{e}{c}A(q)\right)B_\lambda(x, p) \\ \mathcal{H}A_\lambda(\vec{x} - \vec{q}') &= \int dq e^{i\frac{e}{c}A(q')\cdot q}E_\lambda(\vec{q}' - q)A_\lambda(\vec{x} - \vec{q}) \end{aligned} \quad (43)$$

and the lattice Weyl transform of any operator, A_{op} , is

$$a_{\lambda\lambda'}(p, q) = \int d\vec{v} e^{\frac{i}{\hbar}\vec{p}\cdot\vec{v}} \left\langle A_\lambda\left(\vec{q} - \frac{1}{2}\vec{v}\right) \left| A_{op} \right| A_{\lambda'}\left(\vec{q} + \frac{1}{2}\vec{v}\right) \right\rangle \quad (44)$$

The Weyl transform of the Hamiltonian operator is easily calculated using Eq. (43) and Eq. (44). The reader is referred to Ref. ^(11,12) for details of the derivation. Applying Eq. (44) to the even form of the Dirac Hamiltonian, we have

$$\begin{aligned} h'_B(\vec{p}, \vec{q})_{\lambda\lambda'} &= \int d\vec{v} \, e^{\frac{i}{\hbar}\vec{p}\cdot\vec{v}} \left\langle A_\lambda \left(\vec{q} - \frac{1}{2}\vec{v} \right) \left| \mathcal{H}'_B \right| A_{\lambda'} \left(\vec{q} + \frac{1}{2}\vec{v} \right) \right\rangle \\ &= \int d\vec{v} \, \exp \left[\frac{i}{\hbar} \left(p - \frac{e}{c} A(q) \right) \cdot v \right] \tilde{E}_\lambda(v; B) \delta_{\lambda\lambda'} \\ &= E_\lambda \left(\vec{p} - \frac{e}{c} A(q); B \right) \delta_{\lambda\lambda'} \end{aligned}$$

D. The function $E_\lambda(\vec{p} - \frac{e}{c}A(q); B)\delta_{\lambda\lambda'}$

The function $E_\lambda(\vec{p} - \frac{e}{c}A(q); B)$ is the Weyl transform of $\beta[\mathcal{H}^2]^{\frac{1}{2}}$, where the matrix β served to designate the four bands. In order to calculate χ we only need the knowledge of $E_\lambda(\vec{p} - \frac{e}{c}A(q); B)$ as an expansion up to second order in the coupling constant e and after a change of variable [this is effected by setting $A(q) = 0, p = \hbar k$ in the expansion], we obtain the expression of $E_\lambda(\vec{p} - \frac{e}{c}A(q); B)|_{A(q)=0}$, where the dependence in the field B is beyond the vector potential,

$$E_\lambda(\vec{k}; B) = E_\lambda(\vec{k}; B) + BE_\lambda^{(1)}(\vec{k}) + B^2E_\lambda^{(2)}(\vec{k}) + \dots$$

The function $E_\lambda(\vec{p} - \frac{e}{c}A(q); B)|_{A(q)=0}$ which includes the anomalous magnetic moment of the electron is obtained as

$$\begin{aligned} E_\lambda(k; B) = & \beta \left\{ E - \frac{ec}{2E} \vec{L}_{c.m.} \cdot \vec{B} - \frac{(1+\lambda')}{2E} e\hbar c \vec{\sigma} \cdot \vec{B} - \frac{(1+\lambda')^2}{8E^3} \left(e\hbar c \vec{\sigma} \cdot \vec{B} \right)^2 \right. \\ & \left. + \frac{(e\hbar c)^2 \epsilon^2}{8E^5} B^2 \left[1 + \left(\frac{\lambda' E}{mc^2} \right)^2 \right] + O(e^3) \right\} \end{aligned}$$

where

$$\begin{aligned} \vec{L}_{c.m.} &= \beta \left(\frac{\lambda' \hbar}{mc} \right) \vec{\sigma} \times \vec{p} \\ \epsilon^2 &= m^2 c^4 + c^2 \hbar^2 k_z^2 \\ E(\vec{k}) &= \left[m^2 c^4 + c^2 \hbar^2 k^2 \right]^{\frac{1}{2}} \end{aligned}$$

The term, $\vec{L}_{c.m.}$, is a magnetodynamic effect, i.e., due to hidden average angular momentum $\vec{L}_{c.m.}$ of a moving electron. Thus, the introduction of the Pauli anomalous term in \mathcal{H} at

the outset endows a rigid-body behavior to the electron, and its angular momentum about the origin \vec{L}_0 is

$$\vec{L}_0 = \vec{L}_{MO} + \vec{L}_{c.m.}$$

where \vec{L}_{MO} is the angular momentum about the origin of the system of charge concentrated as a point at the center of mass and $\vec{L}_{c.m.}$ is the average angular momentum of the system, as a spread-out distribution of charge about the center of mass. Thus,

$$\begin{aligned}\vec{L}_0 &= \vec{q} \times \vec{p} + \left\langle \sum_i \vec{r}_i \times \vec{p}_i \right\rangle \\ \left\langle \sum_i \vec{r}_i \times \vec{p}_i \right\rangle &= \beta \left(\frac{\lambda' \hbar}{mc} \right) \vec{\sigma} \times \vec{p}\end{aligned}$$

$$\begin{aligned}M &= - \left[2E_\lambda^{(2)}(\vec{k}) B \right]_{sp} \\ &= - \frac{(e\hbar c)^2 \epsilon^2}{4[E_\lambda(\vec{k})]^5} \left[1 + \left(\frac{\lambda' E}{mc^2} \right)^2 \right] B\end{aligned}\tag{45}$$

The induced magnetic moment due to a distribution of electric charge is

$$M = - \frac{Be^2 \langle r^2 \rangle}{4mc^2}\tag{46}$$

where $\langle r^2 \rangle$ is the average of the square of the spatial spread of the distribution normal to the magnetic field. Equating Eqs. (45) with (46) we obtain

$$\langle r^2 \rangle = \frac{mc^2 (\hbar c)^2 \epsilon^2}{[E_\lambda(k)]^5} \left[1 + \left(\frac{\lambda' E}{mc^2} \right)^2 \right]\tag{47}$$

For positive energy states $E_\lambda(k) = (c^2 \hbar^2 k^2 + m^2 c^4)^{\frac{1}{2}}$ and in the nonrelativistic limit, Eq. (47) reduces to

$$\langle r^2 \rangle = (1 + \lambda'^2) \left(\frac{\hbar}{mc} \right)^2$$

and thus the effective spread of the electron at rest, and for $\lambda' = 0$, is precisely equal to the Compton wavelength.

E. Magnetic Susceptibility of Dirac Fermions

The magnetic susceptibility is given by

$$\begin{aligned}\chi = & -\frac{1}{48\pi^3} \left(\frac{e}{\hbar c}\right)^2 \sum_{\lambda} \int d\vec{k} \left\{ \frac{\partial^2 E_{\lambda}(\vec{k}; 0)}{\partial k_x^2} \frac{\partial^2 E_{\lambda}(\vec{k}; 0)}{\partial k_y^2} - \left(\frac{\partial^2 E_{\lambda}(\vec{k}; 0)}{\partial k_x \partial k_y} \right)^2 \right\} \frac{\partial f(E_{\lambda})}{\partial E_{\lambda}} \\ & - \left(\frac{1}{2\pi}\right)^3 \sum_{\lambda} \int d\vec{k} \left[E_{\lambda}^{(1)}(k) \right]^2 \frac{\partial f(E_{\lambda})}{\partial E_{\lambda}} - \left(\frac{1}{2\pi}\right)^3 \sum_{\lambda} \int d\vec{k} 2E_{\lambda}^{(2)}(k) f(E_{\lambda})\end{aligned}$$

Using the following change of variable of integration,

$$(\hbar c)^3 \int d\vec{k} = \int_{-\infty}^{\infty} d\eta \int_0^{2\pi} d\phi E(\vec{k}) dE(\vec{k})$$

where

$$\eta = \hbar c k_z$$

we obtain for the positive energy states the expression for χ which can be divided into more physically meaningful terms as

$$\chi = \chi_{LP} + \chi_P + \chi_{sp} + \chi_g + \chi_{MD}$$

where

$$\chi_{LP} = \frac{1}{24\pi^3} \left(\frac{e}{\hbar c}\right)^2 \int_{-\infty}^{\infty} d\eta \int_{\epsilon}^{\infty} \frac{\epsilon^2}{E^3} \frac{\partial f(E)}{\partial E} dE \quad (48)$$

$$\chi_P = -\frac{(1+\lambda')^2}{8\pi^2} \left(\frac{e}{\hbar c}\right)^2 \int_{-\infty}^{\infty} d\eta \int_{\epsilon}^{\infty} \frac{1}{E} \frac{\partial f(E)}{\partial E} dE \quad (49)$$

$$\chi_{sp} = -\frac{1}{8\pi^2} \left(\frac{e^2}{\hbar c}\right) \int_{-\infty}^{\infty} d\eta \int_{\epsilon}^{\infty} \frac{\epsilon^2}{E^4} \left[1 + \left(\frac{\lambda' E}{mc^2} \right)^2 \right] f(E) dE \quad (50)$$

$$\chi_g = \frac{(1+\lambda')^2}{8\pi^2} \left(\frac{e^2}{\hbar c}\right) \int_{-\infty}^{\infty} d\eta \int_{\epsilon}^{\infty} \frac{f(E)}{E^2} dE \quad (51)$$

$$\chi_{MD} = -\frac{\lambda'^2}{8\pi^2} \left(\frac{e^2}{\hbar c}\right) \int_{-\infty}^{\infty} d\eta \int_{\epsilon}^{\infty} \frac{(E^2 - \epsilon^2)}{(mc^2)^2 E} \frac{\partial f(E)}{\partial E} dE \quad (52)$$

where

$$\begin{aligned}\left(\frac{ec}{2E} \vec{L}_{c.m.} \right)_z^2 &= \left(\frac{\lambda' e \hbar c}{2mc^2} \right) \frac{(E^2 - \epsilon^2)}{E^2} \\ \vec{B} &= B \frac{\vec{z}}{|\vec{z}|}\end{aligned}$$

The total susceptibility for the positive energy states is

$$\chi = \frac{1}{(2\pi)^2} \left(\frac{e^2}{\hbar c} \right) \left[(1 + \lambda')^2 - \frac{1}{3} \right] \int_0^\infty d\eta \frac{f(\epsilon)}{\epsilon} \frac{1}{(2\pi)^2} \left(\frac{e^2}{\hbar c} \right) \left(\frac{\lambda'}{mc^2} \right)^2 \int_0^\infty d\eta G(\epsilon - \mu) \quad (53)$$

where

$$G(\epsilon - \mu) = k_B T \ln \left\{ 1 + \exp \left[- \frac{(\epsilon - \mu)}{k_B T} \right] \right\} \\ = \int_\epsilon^\infty f(E) dE$$

The contributions of the holes is obtained by replacement of $f(\epsilon)$ and $G(\epsilon - \mu)$ in Eq. (53) by $(1 - f(-\epsilon))$ and $G(\epsilon + \mu)$, respectively.

The relative importance of terms that made up χ at $T = 0$ of Dirac fermions, where n is the electron density, $k_F = (3\pi^2 n)^{\frac{1}{3}}$, $\eta_F = \hbar c k_F$ and $E_F = (\Delta^2 + \eta_F^2)^{\frac{1}{2}}$, is summarized below.

Various Contributions to χ_{Dirac} at $T = 0$	Nonrelativistic, $\frac{\eta_F}{\Delta} \ll 1$	Ultrarelativistic, $\frac{\eta_F}{\Delta} \gg 1$
$\chi_{LP} = -\frac{1}{12\pi^2} \left(\frac{e^2}{\hbar c} \right) \frac{1}{E_F^3} \left(\frac{\eta_F^3}{3} + \Delta^2 \eta_F \right)$	$-\frac{1}{12\pi^2} \left(\frac{e}{mc^2} \right) k_F$	$-\frac{1}{12\pi^2} \left(\frac{e^2}{\hbar c} \right) \frac{1}{3}$
$\chi_P = \frac{1}{4\pi^2} (1 + \lambda')^2 \left(\frac{e^2}{\hbar c} \right) \frac{\eta_F}{E_F}$	$\frac{1}{4\pi^2} (1 + \lambda')^2 \left(\frac{e}{mc^2} \right) k_F$	$\frac{1}{4\pi^2} (1 + \lambda')^2 \left(\frac{e^2}{\hbar c} \right)$
$\chi_{MD} = -\frac{\lambda'^2}{4\pi^2} \left(\frac{e^2}{\hbar c} \right) \frac{1}{\Delta^2} \left[\frac{1}{E_F} \left(\frac{\eta_F^3}{3} + \Delta^2 \eta_F \right) - \eta_F E_F \right]$	$\Rightarrow 0$	$\frac{\lambda'^2}{4\pi^2} \left(\frac{e^2}{\hbar c} \right) \frac{2}{3} \left(\frac{\eta_F}{\Delta} \right)^2$
$\chi_{spread} = -\frac{1}{12\pi^2} \left(\frac{e^2}{\hbar c} \right) \sinh^{-1} \left(\frac{\eta_F}{\Delta} \right) - \chi_{LP}$		$-\frac{1}{12\pi^2} \left(\frac{e^2}{\hbar c} \right) \left[\ln \frac{2\eta_F}{\Delta} - \frac{1}{3} \right]$
$-\frac{\lambda'^2}{4\pi^2} \left(\frac{e^2}{\hbar c} \right) \frac{1}{\Delta^2} \left[\frac{\eta_F(\eta_F^2 + \Delta^2)^{\frac{1}{2}}}{2} + \frac{\Delta^2}{2} \sinh^{-1} \left(\frac{\eta_F}{\Delta} \right) \right]$		
$+\frac{\lambda'^2}{4\pi^2} \left(\frac{e^2}{\hbar c} \right) \frac{1}{E_F} \left(\frac{\eta_F^3}{3} + \Delta^2 \eta_F \right) \frac{1}{\Delta^2}$	$\Rightarrow 0$	$-\frac{\lambda'^2}{4\pi^2} \left(\frac{e^2}{\hbar c} \right) \left[\frac{1}{6} \left(\frac{\eta_F}{\Delta} \right)^2 + \frac{1}{2} \ln \frac{2\eta_F}{\Delta} \right]$
$\chi_g = \frac{1}{4\pi^2} (1 + \lambda')^2 \left(\frac{e^2}{\hbar c} \right) \left[\sinh^{-1} \left(\frac{\eta_F}{\Delta} \right) - \frac{\eta_F}{E_F} \right]$	$\Rightarrow 0$	$\frac{1}{4\pi^2} (1 + \lambda')^2 \left(\frac{e^2}{\hbar c} \right) \left[\ln \frac{2\eta_F}{\Delta} - 1 \right]$

F. Displacement Operator under Uniform High External Electric Fields

To complement Sec. V C, we give the translation operator for uniform electric field case, $\mathcal{H} = \mathcal{H} - e\vec{F} \cdot \vec{x}$. We have for the displacement operator, $T_E(q)$, obeying the relation,

$$i\hbar \dot{T}_E(q) = [T_E(q), \mathcal{H}] \\ \dot{T}_E(q) = \frac{ie}{\hbar} F \cdot q T_E(q)$$

Therefore

$$T_E(q) = C_0(q, \tau) \exp \left(\frac{ie}{\hbar} F t \cdot q \right)$$

where $C_0(q, \tau)$ is an operator which do not depend explicitly on time, t . $T_E(q)$, being a displacement operator in space and time lead us to write the operator

$$C_0(q, \tau) = \exp \left(q \cdot \frac{\partial}{\partial r} + \tau \frac{\partial}{\partial t} \right)$$

$T_E(q)$ plays critical role similar to $T_M(q)$ for establishing the phase space quantum transport dynamics at very high electric fields, where we consider realistic transport problems as time-dependent many-body problems. For zero field case we are dealing with biorthogonal Wannier functions and Bloch functions because the Hamiltonian is no longer Hermetian due to the presence of energy variable, z , in the self-energy. This means that $[T_E(q), \mathcal{H}]$ is diagonal in the bilinear expansion if $T_E(q)$ is diagonal. The eigenfunction of the ‘lattice’ translation operator $T_E(q)$ must then be labeled by a wavenumber \vec{k} which is varying in time as

$$\vec{k} = \vec{k}_0 + \frac{e\vec{F}}{\hbar}t$$

and \mathcal{H} is also diagonal in \vec{k} . Similarly, the energy variable, z , in the Hamiltonian must also vary as

$$z = z_0 + e\vec{F} \cdot \vec{q}$$

Similar developments for translationally invariant many-body system subjected to a uniform electric field allows us to define the corresponding electric Bloch functions and electric Wannier functions, in a unifying manner for both magnetic and electric fields. This electric-field version allows us to derive the quantum transport equation of the particle density at very high electric fields. This will be discussed in another communication dealing with quantum transport in many-body systems.

VI. MAGNETIC SUSCEPTIBILITY OF MANY-BODY SYSTEMS IN A UNIFORM FIELD

Here, we shall see that symmetry arguments enable us to generalize, in a unified manner, the derivation of χ for noninteracting to that of interacting Fermi systems possessing translational symmetry.⁶²

The reduced one-particle Schrodinger equation of a many-body system in the presence of a uniform magnetic field is defined by

$$[\mathcal{H}_0 + \Sigma(z)]\phi(z) = E(z)\phi(z), \tag{54}$$

where $\Sigma(z)$ is the nonlocal energy-dependent (z is the energy variable) complex quantity called the self-energy operator. \mathcal{H}_0 is the non-interacting Hamiltonian in a magnetic field. In the absence of spin-orbit coupling, this is given by

$$\mathcal{H}_0 = \frac{1}{2m} \left(\frac{\hbar}{i} \nabla_{\vec{r}} - \frac{e}{c} \vec{A}(\vec{r}) \right)^2 + V(\vec{r}) - g\mu_B S_z B \quad (55)$$

$V(\vec{r}) + \Sigma(z)$ represents the effective potential which is a non-Hermitian operator leading to the use of biorthogonal eigenfunctions, with the dual sets obtained from the eigenfunctions of $\mathcal{H}_0 + \Sigma(z)$ and its adjoint. In the presence of spin-orbit coupling, we have

$$\mathcal{H}_0 = \frac{1}{2m} \left(\frac{\hbar}{i} \nabla_{\vec{r}} - \frac{e}{c} \vec{A}(\vec{r}) - \frac{\mu_B}{c} \sigma \times \nabla V(r) \right)^2 - g\mu_B \sigma \cdot B + V(r) \quad (56)$$

which gives the spin-orbit interaction correctly to order $\frac{e}{mc^2}$.⁵⁴ In the presence of inversion and time reversal symmetry the eigenfunctions are spinors and so are the magnetic Wannier function and magnetic Bloch function themselves.

A. The Crystalline Effective Hamiltonian

We will transform the many-body effective-Hamiltonian operator $\mathcal{H}_0 + \Sigma(z)$ to an effective Hamiltonian expressed in terms of the crystal-momentum operator, \vec{P} , and the lattice-position operator, \vec{Q} . This is done through the lattice Weyl-Wigner formalism of the quantum dynamics of solids.² We have,

$$\begin{aligned} \mathcal{H}_{eff}(\vec{P}, \vec{Q}, z) = & (N\hbar^3)^{-2} \sum_{\vec{p}, \vec{q}, \lambda, \lambda', \vec{u}, \vec{v}} H_{\lambda\lambda'} \left(\vec{p} - \frac{e}{c} \vec{A}(\vec{q}); B, z \right) \exp \left(\frac{2i}{\hbar} (\vec{p} - \vec{P}) \cdot \vec{v} \right) \\ & \times \exp \left(\frac{2i}{\hbar} (\vec{q} - \vec{Q} - \vec{v}) \cdot \vec{u} \right) \Omega_{\lambda\lambda'}, \end{aligned} \quad (57)$$

where

$$H_{\lambda\lambda'} \left(\vec{p} - \frac{e}{c} \vec{A}(\vec{q}); B, z \right) = \sum_{\vec{v}} e^{(\frac{2i}{\hbar}) \vec{p} \cdot \vec{v}} \langle \vec{q} - \vec{v}, \lambda | \mathcal{H}_0 + \Sigma(z) | \vec{q} + \vec{v}, \lambda' \rangle \quad (58)$$

or in the presence of spin-orbit interaction, this is given by,

$$H_{\lambda\lambda'} \left(\vec{p} - \frac{e}{c} \vec{A}(\vec{q}) - \frac{\mu_B}{c} \sigma \times \nabla V(q); B, z \right) = \sum_{\vec{v}} e^{(\frac{2i}{\hbar}) \vec{p} \cdot \vec{v}} \langle \vec{q} - \vec{v}, \lambda | \mathcal{H}_0 + \Sigma(z) | \vec{q} + \vec{v}, \lambda' \rangle \quad (59)$$

where

$$\Omega_{\lambda\lambda'} = \sum_{\vec{q}} |\vec{q}, \lambda\rangle \langle \vec{q}, \lambda'| = \sum_{\vec{p}} |\vec{p}, \lambda\rangle \langle \vec{p}, \lambda'| \quad (60)$$

and the $|\vec{q}, \lambda\rangle$ or $|\vec{p}, \lambda\rangle$ are considered spinors for each band index λ in the case with spin-orbit interaction. We expand the eigensolutions of (57) in terms of the complete set of magnetic Wannier functions or of magnetic Bloch functions of non-interacting system, \mathcal{H}_0 ,

$$\phi(\vec{r}, z) = \sum_{\vec{p}, \lambda} f_\lambda(\vec{p}, z) |\vec{p}, \lambda\rangle \quad (61)$$

$$\phi(\vec{r}, z) = \sum_{\vec{q}, \lambda} f_\lambda(\vec{q}, z) |\vec{q}, \lambda\rangle \quad (62)$$

an equivalent eigenvalue problem is obtained for the coefficients $f_\lambda(\vec{q}, z)$. In \vec{q} -space this is,

$$\sum_{\lambda'} W_{\lambda\lambda'}(\vec{\pi}; B, z) f_{\lambda'}(\vec{q}, z) = E(z) f_\lambda(\vec{q}, z) \quad (63)$$

and the corresponding eigenvalue equation in \vec{p} -space

$$\sum_{\lambda'} W_{\lambda\lambda'}(\vec{\pi}; B, z) f_{\lambda'}(\vec{p}, z) = E(z) f_\lambda(\vec{p}, z) \quad (64)$$

where

$$W_{\lambda\lambda'}(\vec{\pi}; B, z) = (N\hbar^3)^{-1} \sum_{\vec{p}', \vec{v}} H_{\lambda\lambda'}(\vec{p}'; B, z) \exp\left(\frac{2i}{\hbar}(\vec{p}' - \vec{\pi}) \cdot \vec{v}\right) \quad (65)$$

$$\vec{\pi} = \begin{cases} \frac{\hbar}{i} \nabla_{\vec{q}} - \frac{e}{c} \vec{A}(\vec{q}) & \text{in } \vec{q} \text{ space} \\ \vec{p} + \frac{e}{c} \vec{A}\left(\frac{\hbar}{i} \nabla_{\vec{p}}\right) & \text{in } \vec{p} \text{ space} \end{cases} \quad (66)$$

Since $W_{\lambda\lambda'}(\vec{\pi}; B, z)$ is a non-Hermitian operator, one also needs to solve the adjoint problem, either in q -space or \vec{p} -space,

$$\sum_{\lambda'} W_{\lambda'\lambda}^*(\vec{\pi}; B, z) e_{\lambda'}(\vec{q}, z) = E^*(z) e_\lambda(\vec{q}, z) \quad (67)$$

$$\sum_{\lambda'} W_{\lambda'\lambda}^*(\vec{\pi}; B, z) e_{\lambda'}(\vec{p}, z) = E^*(z) e_\lambda(\vec{p}, z) \quad (68)$$

$W_{\lambda\lambda'}(\vec{\pi}; B, z)$ may be viewed as a generalized Hamiltonian of the Dirac type occurring in the relativistic quantum theory of electrons. Since we are using magnetic Wannier functions and magnetic Bloch functions of the noninteracting Bloch electrons in a uniform magnetic field as basis states, \mathcal{H}_0 is diagonal in band indices and we may write

$$W_{\lambda\lambda'}(\vec{\pi}; B, z) = W_0(\vec{\pi}; B, z) \delta_{\lambda\lambda'} + \Sigma_{\lambda\lambda'}(\vec{\pi}; B, z), \quad (69)$$

where $W_0(\vec{\pi}; B, z)_\lambda$ is the effective magnetic Hamiltonian, belonging to the band, λ , of noninteracting Bloch electrons in a uniform magnetic field. In the case with spin-orbit

interaction, $W_{\lambda\lambda'}(\vec{\pi}; B, z)$ is a spinor for each band index. Just like the relativistic Dirac Hamiltonian, $W_{\lambda\lambda'}(\vec{\pi}; B, z)$ can be transformed into an *even form*, that is without any off-diagonal terms through the technique of successive transformation as defined below.

The eigenfunctions $f_{\lambda'}(\vec{q}, z)$ of $W_{\lambda\lambda'}(\vec{\pi}; 0, z) \equiv \mathcal{H}_{\lambda\lambda'}^{(0)}$ and those of its adjoint define a similarity transformation which diagonalizes $\mathcal{H}_{\lambda\lambda'}^{(0)}$. We have

$$U^{-1}H^0U = \tilde{H}_\lambda^0\delta_{\lambda\lambda'} \quad (70)$$

where the matrix of U is given by f_{ij} , where f_{ij} denotes the i th component of an eigenvector belonging to the j th eigenvalue of the matrix $\mathcal{H}_{\lambda\lambda'}^{(0)}$. The matrix of U^{-1} is the matrix formed by e_{ji}^* , where e_{ji} is the i th component of the j th eigenvector of the adjoint matrix. U and U^{-1} also determine the transformation from the Wannier function and Bloch function of non-interacting Bloch electrons to the Wannier function and Bloch function of interacting Bloch electrons, which are, in general, energy dependent and biorthogonal. Denoting these by $|\vec{q}, \lambda, z\rangle$ and $|\vec{p}, \lambda, z\rangle$, we have

$$|\vec{p}, \lambda, z\rangle = \sum_i f_{i\lambda}(\vec{p}, z) |\vec{p}, i\rangle \quad (71)$$

$$\langle \vec{p}, \lambda, z | = \sum_i e_{\lambda i}^*(\vec{p}, z) \langle \vec{p}, i | \quad (72)$$

and hence we have,

$$|\vec{q}, \lambda, z\rangle = (N\hbar^3)^{-\frac{1}{2}} \sum_{\vec{p}} e^{\frac{i}{\hbar}(\vec{p}\cdot\vec{q})} |\vec{p}, \lambda, z\rangle \quad (73)$$

$$\langle \vec{q}, \lambda, z | = \sum_{\vec{p}} e^{\frac{i}{\hbar}(\vec{p}\cdot\vec{q})} \langle \vec{p}, \lambda, z | \quad (74)$$

In terms of these basis states, $\tilde{H}_\lambda^0(\vec{p}, z)\delta_{\lambda\lambda'}$, is given by

$$\tilde{H}_\lambda^0(\vec{p}, z) = \sum_{\vec{v}} e^{\frac{2i}{\hbar}\vec{p}\cdot\vec{v}} \langle \vec{q} - \vec{v}, \lambda, z | [\mathcal{H}_0 + \Sigma(z)] | \vec{q} + \vec{v}, \lambda, z \rangle \quad (75)$$

or equivalently,

$$\tilde{H}_\lambda^0(\vec{p}, z) = \sum_{\vec{u}} e^{\frac{2i}{\hbar}\vec{q}\cdot\vec{u}} \langle \vec{p} + \vec{u}, \lambda, z | [\mathcal{H}_0 + \Sigma(z)] | \vec{p} - \vec{u}, \lambda, z \rangle \quad (76)$$

The one-particle energy z_λ belonging to the band index λ is, in the quasiparticle picture given as usual by the solution of

$$z_\lambda - \tilde{H}_\lambda^0(\vec{p}, z_\lambda) = 0 \quad (77)$$

which is doubly degenerate in the case with spin-orbit interaction.

B. Removal of Interband Terms of \mathcal{H}_{eff} in a Magnetic Field

The basic idea is that instead of transforming the operator $W_{\lambda\lambda'}(\vec{\pi}; B, z)$ to an even form directly, one tries to transform the lattice Weyl transform of $\mathcal{H}_{eff}(\vec{P}, \vec{Q})$ into an even form. The power and advantage of this approach lies in being able to deal with ordinary c -numbers instead of quantum mechanical operators. To diagonalize the operator $W_{\lambda\lambda'}(\vec{\pi}; B, z)$ or transform to an *even form*, we seek a transformation S_{op} such that

$$S_{op}^{-1} \mathcal{H}_{eff} S_{op} \rightleftharpoons \tilde{H}_\lambda \left(\vec{p} - \frac{e}{c} \vec{A}(\vec{q}); B, z \right) \delta_{\lambda\lambda'} \quad (78)$$

where \rightleftharpoons indicates the one-to-one Weyl correspondence between operator and its lattice Weyl transform. The $\delta_{\lambda\lambda'}$ includes the diagonalization with respect to the Kramer's conjugate or degenerate bands, analogous to the transformation of the Dirac relativistic Hamiltonian to an even form. The transformed effective Hamiltonian operator $\bar{W}_\lambda(\vec{\pi}; B, z)$ for each band index λ is thus given using the diagonalized Weyl transform \tilde{H}_λ as

$$\bar{W}_\lambda(\vec{\pi}; B, z) = (N\hbar^3)^{-1} \sum_{\vec{p}', \vec{v}} \tilde{H}_\lambda(\vec{p}'; B, z) \exp \left(\frac{2i}{\hbar} (\vec{p}' - \vec{\pi}) \cdot \vec{v} \right) \quad (79)$$

The Weyl transform of a product of three operators is given by the following expression,

$$\begin{aligned} & S_{op}^{-1} \mathcal{H}_{eff} S_{op} \\ & \rightleftharpoons \exp \left[\frac{i\hbar e B}{2c} \left(\frac{\partial^{(a)}}{\partial \vec{k}_x} \frac{\partial^{(b)}}{\partial \vec{k}_y} - \frac{\partial^{(a)}}{\partial \vec{k}_y} \frac{\partial^{(b)}}{\partial \vec{k}_x} + \frac{\partial^{(a)}}{\partial \vec{k}_x} \frac{\partial^{(c)}}{\partial \vec{k}_y} - \frac{\partial^{(a)}}{\partial \vec{k}_y} \frac{\partial^{(c)}}{\partial \vec{k}_x} + \frac{\partial^{(b)}}{\partial \vec{k}_x} \frac{\partial^{(c)}}{\partial \vec{k}_y} - \frac{\partial^{(b)}}{\partial \vec{k}_y} \frac{\partial^{(c)}}{\partial \vec{k}_x} \right) \right] \\ & \times S^{-1(a)}(\vec{k}, B, z) H^{(b)}(\vec{k}, B, z) S^{(c)}(\vec{k}, B, z) \end{aligned} \quad (80)$$

The procedure is to diagonalize the lattice Weyl transform of \mathcal{H} , by means of successive similarity transformations

$$S_{op} = \prod_{i=1}^{\infty} S_{op}^0 e^{G_{op}^{(i)}} \quad (81)$$

To find S_{op}^0 we expand $H(\vec{k}, B, z)$ in powers of B ,

$$H(\vec{k}, B, z) = H^0(\vec{k}, z) + B H^{(1)}(\vec{k}, z) + \dots \quad (82)$$

and require that the zero-order term on the right-hand side of Eq. (80) be diagonal. Denoting the matrix which diagonalizes $H^0(\vec{k}, z)$ by $U(\vec{k}, z)$ we have

$$U^{-1}(\vec{k}, z) H^0(\vec{k}, z) U(\vec{k}, z) = \tilde{H}_\lambda^0(\vec{k}, z) \delta_{\lambda\lambda'} \quad (83)$$

Equation (83) is a pure matrix diagonalization problem and was already solved above for the zero-field case. There, we have assumed that the eigenvalues of $H^0(\vec{k}, z)$ are nondegenerate; the resulting eigenvectors of $H^0(\vec{k}, z)$ and those of its adjoint define a similarity transformation from Wannier function and Bloch function for $\Sigma = 0$ to the Wannier function and Bloch function for $\Sigma \neq 0$, which are, in general, energy dependent and biorthogonal.

C. Iterative Solution for a Unitary $U(\vec{k}, z)$

The operator corresponding to $U(\vec{k}, z)$, Eq. (83), is, however, a similarity transformation only for the zero-field case. Thus, setting $\mathcal{H}_{eff} = 1$ in Eq. (80), we have,

$$\{U_{op}^{-1}U_{op}\} \rightleftharpoons \exp \left[\frac{i\hbar eB}{2c} \left(\frac{\partial^{(a)}}{\partial \vec{k}_x} \frac{\partial^{(b)}}{\partial \vec{k}_y} - \frac{\partial^{(a)}}{\partial \vec{k}_y} \frac{\partial^{(b)}}{\partial \vec{k}_x} \right) \right] U_{op}^{-1(a)}(\vec{k}, z) U_{op}^{(b)}(\vec{k}, z) \quad (84)$$

where $\{U_{op}^{-1}U_{op}\}$ indicates that the product is not to be interpreted as exact product of an operator and its inverse. However, $\{U_{op}^{-1}U_{op}\}$ can be made equal to unity up to an arbitrary order in the magnetic field strength B by means of successive multiplication by exponential operators on the left- and right-hand sides. We have

$$U_{op}^{-1(n)}U_{op}^{(n)} = \prod_{i=n}^1 e^{g_{op}^{(i)}} \{U_{op}^{-1}U_{op}\} \prod_{i=1}^n e^{g_{op}^{(i)}} = 1 + O(B^n) \quad (85)$$

where each successive $e^{g_{op}^{(i)}}$ is so chosen so as to make the product unity up to order i in the magnetic field strength.

To prove this, we need the expression for the lattice Weyl transform of an arbitrary operator A_{op} raised to any power n . For problems involving uniform magnetic field and possessing translational symmetry, this can be written analogous to Eq. (80) as

$$A_{op}^n \rightleftharpoons \cos \left[\frac{e\hbar B}{2c} \sum_{\substack{j,k=1 \\ j < k}}^n \left(\frac{\partial^{(j)}}{\partial \vec{k}_x} \frac{\partial^{(k)}}{\partial \vec{k}_y} - \frac{\partial^{(j)}}{\partial \vec{k}_y} \frac{\partial^{(k)}}{\partial \vec{k}_x} \right) \right] \frac{1}{2} \left(\prod_{l=1}^n A^{(l)}(\vec{k}; B) + \prod_{l=n}^1 A^{(l)}(\vec{k}; B) \right) \quad (86)$$

The lattice Weyl transform of an exponential operator $\exp(g_{op}^{(i)})$ can therefore be expressed as

$$\begin{aligned} \exp(g_{op}^{(i)}) &\rightleftharpoons \exp[g^{(i)}(\vec{k}; B)] + R \\ &= 1 + g^{(i)}(\vec{k}; B) + \dots + R \end{aligned} \quad (87)$$

where R represents the remaining terms and $g_{op}^{(i)} \rightleftharpoons g^{(i)}(\vec{k}; B)$. A complete iteration procedure for obtaining S_{op} in Eq. (81), up to an arbitrary order in B can now be defined.

Let us write Eq. (84) as

$$\{U_{op}^{-1}U_{op}\} \rightleftharpoons 1 + BS^{(1)}(\vec{k}, z) + B^2S^{(2)}(\vec{k}, z) + \dots \quad (88)$$

where the explicit dependence of B comes from the exponential ‘‘Poisson-bracket operator’’ in (84). We choose $g_{op}^{(1)} \rightleftharpoons -\frac{1}{2}BS^{(1)}(\vec{k}, z)$, obtaining

$$e^{g_{op}^{(1)}}\{U_{op}^{-1}U_{op}\}e^{g_{op}^{(1)}} \rightleftharpoons 1 + B^2S^{(2)}(\vec{k}, z) + \Delta_R^{(1)} \quad (89)$$

We next choose, $g_{op}^{(2)} = -\frac{1}{2}B^2S^{(2)}(\vec{k}, z)$ resulting in

$$e^{g_{op}^{(2)}}e^{g_{op}^{(1)}}\{U_{op}^{-1}U_{op}\}e^{g_{op}^{(1)}}e^{g_{op}^{(2)}} \rightleftharpoons 1 + B^3S^{(3)}(\vec{k}, z) + \Delta_R^{(2)} \quad (90)$$

In general order n , we have

$$U_{op}^{-1(n)}U_{op}^{(n)} \rightleftharpoons 1 + B^{n+1}S^{(n+1)}(\vec{k}, z) + \Delta_R^{(n)} \quad (91)$$

and $g_{op}^{(n)}$ can be chosen such that $g_{op}^{(n)} = -\frac{1}{2}B^nS^{(n)}(\vec{k}, z)$. This completes the proof.

D. Iterative Removal of Interband Terms of \mathcal{H}_{eff}

We now proceed to the diagonalization of \mathcal{H}_{eff} , Eq. (78). Let us write the expression containing the zero-order diagonal, $\tilde{H}_\lambda^0(\vec{k}, z)\delta_{\lambda\lambda'}$, as

$$\left(S_{op}^0\right)^{-1}\mathcal{H}_{eff}S_{op}^0 \rightleftharpoons \tilde{H}_\lambda^0(\vec{k}, z)\delta_{\lambda\lambda'} + BH^{(1)}(\vec{k}, z)_{\lambda\lambda'} + \Delta_R \quad (92)$$

Note that for even function of coordinates, linear momentum, the velocity, the spin-orbit interaction, and a symmetrized product of an even number of linear momenta do *not* couple the Kramer’s conjugates or doubly degenerate states of the same energy. Thus, any other operators which couple the Kramer’s conjugate states must result in spin splitting of the doubly degenerate states over the whole Brillouin zone. By virtue of time reversal and inversion symmetry, we assume that $\tilde{H}_\lambda^0(\vec{k}, z)\delta_{\lambda\lambda'} \equiv \tilde{H}_\lambda^0(\vec{k}, z)\delta_{\lambda\lambda'}\delta_{\lambda,\sigma\sigma'}\delta_{\lambda',\sigma\sigma'}$, indicating that for $\Sigma \neq 0$, the Hamiltonian $\tilde{H}_\lambda^0(\vec{k}, z)\delta_{\lambda\lambda'}$ in the absence of magnetic field is also brought to an even form in terms of the doubly degenerate states in the case with spin-orbit interaction. In the presence of magnetic field the doubly degenerate states are spin-split. Our task for

$\Sigma \neq 0$ is to diagonalize \mathcal{H}_{eff} by method of successive similarity transformation starting with S_{op}^0 , Eq. (81). We will do this for a many-body system, $\Sigma \neq 0$, in a magnetic field.

Let us define “odd” and “even” operators and matrices. An even matrix is a diagonal matrix and the corresponding operator is called an even operator. An odd matrix and its corresponding operator is one where all diagonal (intraband) elements are zeros. Even operators and matrices commute, products of even matrices are even, whereas products of even and odd are odd. The zero-order term on the right-hand side of Eq. (92) is even, the remaining terms may be written as a sum of even and odd matrices. Therefore an iterative procedure to diagonalize \mathcal{H}_{eff} involves removing odd terms on the right-hand side of Eq. (92) up to arbitrary orders in the magnetic field strength. Odd terms in Eq. (92) correspond to the presence of interband terms (including the Kramer’s conjugate bands in the case with spin-orbit interaction which are coupled in a magnetic field) in the effective Hamiltonian and its Weyl transform.

First we choose $G_{op}^{(1)} \rightleftharpoons G^{(1)}(\vec{k}, B, z)$ such that

$$\left[G^{(1)}(\vec{k}, B, z), \tilde{H}^0(\vec{k}, z) \right] = BH_{odd}^{(1)}(\vec{k}, z), \quad (93)$$

where $H_{odd}^{(1)}(\vec{k}, z)$ is the odd part of $H^{(1)}(\vec{k}, z)$ in Eq. (92). Then we have

$$e^{-G_{op}^{(1)}} \left[(S_{op}^0)^{-1} \mathcal{H}_{eff} S_{op}^0 \right] e^{G_{op}^{(1)}} \rightleftharpoons \tilde{H}_\lambda^0(\vec{k}, z) \delta_{\lambda\lambda'} + BH_{even}^{(1)} + B^2 \left[H_{odd}^{(2)} + H_{even}^{(2)} \right] + \Delta_R^{(1)} \quad (94)$$

showing that the right-hand side of Eq. (94) is even up to order B . Since $\tilde{H}^0(\vec{k}, z)$ is even, $G^{(1)}(\vec{k}, B, z)$ can be chosen odd. Its matrix elements is related to that of $H_{odd}^{(1)}(\vec{k}, z)$ and $\tilde{H}^0(\vec{k}, z)$. For $\tilde{H}_j^0(\vec{k}, z) \neq \tilde{H}_i^0(\vec{k}, z)$, this is given by the relation

$$G_{ij}^{(1)}(\vec{k}, B, z) = \begin{cases} B \left[H_{odd}^{(1)}(\vec{k}, z) \right]_{ij} \left[\tilde{H}_j^0(\vec{k}, z) - \tilde{H}_i^0(\vec{k}, z) \right]^{-1} & i \neq j \\ 0 & i = j \end{cases} \quad (95)$$

For the Kramer’s conjugate orthogonal states, $\tilde{H}_i^0(\vec{k}, z)_\sigma = \tilde{H}_i^0(\vec{k}, z)_{\sigma'}$, we asume that the matrix $H_{odd}^{(1)}(\vec{k}, z)_{\sigma\sigma'}$ for each band is of the form of $i\sigma_y$ where σ_y is the Pauli matrix, $\sigma_y = \begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix}$. This form can be obtained by proper choice of the phase of one of the Kramer’s conjugate orthogonal states. Then for the spinor we have from $\left[G_i^{(1)}(\vec{k}, B, z), \tilde{H}^0(\vec{k}, z) \right] = BH_{odd}^{(1)}(\vec{k}, z)_i$, the following relation

$$G_i^{(1)}(\vec{k}, B, z) = \frac{BH_{odd}^{(1)}(\vec{k}, z)_i}{\tilde{H}_i^0(\vec{k}, z)} \quad (96)$$

as the proper choice for the spinor $G_i^{(1)}(\vec{k}, B, z)$ for the each band index i . The procedure can now be reiterated, choosing $G_{op}^{(2)} \rightleftharpoons G^{(2)}(\vec{k}, B, z)$ such that

$$\left[G^{(2)}(\vec{k}, B, z), \tilde{H}^0(\vec{k}, z) \right] = B^2 {}^{(1)}H_{odd}^{(2)}(\vec{k}, z) \quad (97)$$

resulting in

$$\begin{aligned} e^{-G_{op}^{(2)}} e^{-G_{op}^{(1)}} \left[(S_{op}^0)^{-1} \mathcal{H}_{eff} S_{op}^0 \right] e^{G_{op}^{(1)}} e^{G_{op}^{(2)}} \rightleftharpoons \tilde{H}_\lambda^0(\vec{k}, z) \delta_{\lambda\lambda'} + B H_{even}^{(1)} + B^2 H_{even}^{(2)} \\ + B^3 \left[H_{even}^{(3)} + H_{odd}^{(3)} \right] + \Delta_R^{(2)} \end{aligned} \quad (98)$$

with $G^{(2)}(\vec{k}, B, z)$ given by

$$G_{ij}^{(2)}(\vec{k}, B, z) = \begin{cases} B^2 \left[H_{odd}^{(2)}(\vec{k}, z) \right]_{ij} \left[\tilde{H}_j^0(\vec{k}, z) - \tilde{H}_i^0(\vec{k}, z) \right]^{-1} & i \neq j \\ 0 & i = j \end{cases} \quad (99)$$

with the spinor for each band $G_i^{(2)}(\vec{k}, B, z)_{\sigma\sigma'}$ given by

$$G_i^{(2)}(\vec{k}, B, z) = \frac{B H_{odd}^{(2)}(\vec{k}, z)_i}{\tilde{H}_i^0(\vec{k}, z)} \quad (100)$$

In general order n , we can choose $G_{op}^{(n+1)} \rightleftharpoons G^{(n+1)}(\vec{k}, B, z)$ such that if

$$\begin{aligned} \prod_{i=n}^1 e^{-G_{op}^{(i)}} \left[(S_{op}^0)^{-1} \mathcal{H}_{eff} S_{op}^0 \right] \prod_{i=1}^n e^{-G_{op}^{(i)}} \rightleftharpoons \tilde{H}_\lambda^0(\vec{k}, z) \delta_{\lambda\lambda'} + \dots B^n H_{even}^{(n)}(\vec{k}, z) \\ + B^{n+1} \left[H_{odd}^{(n+1)}(\vec{k}, z) + H_{even}^{(n+1)}(\vec{k}, z) \right] + \Delta_R^{(n)} \end{aligned} \quad (101)$$

then we must have $G^{(n+1)}(\vec{k}, B, z)$ given by

$$G_{ij}^{(n+1)}(\vec{k}, B, z) = \begin{cases} B^{n+1} \left[H_{odd}^{(n+1)}(\vec{k}, z) \right]_{ij} \left[\tilde{H}_j^0(\vec{k}, z) - \tilde{H}_i^0(\vec{k}, z) \right]^{-1} & i \neq j \\ 0 & i = j \end{cases} \quad (102)$$

and for the spinor matrix for each band,

$$G_i^{(n+1)}(\vec{k}, B, z) = \frac{B H_{odd}^{(n+1)}(\vec{k}, z)_i}{\tilde{H}_i^0(\vec{k}, z)} \quad (103)$$

The above procedure can, of course, only be guaranteed to converge for very small fields; for calculating the low-field susceptibility the removal of the interband terms up to second order in B is all that is required since higher-order terms do not contribute.

E. Direct Removal of Interband Terms of \mathcal{H} : Magnetic Basis Functions

A lattice Weyl transform of \mathcal{H} which is free of interband terms suggests the existence of magnetic Wannier function and magnetic Bloch function of interacting Bloch electrons in a magnetic field. The starting point in obtaining the lattice Weyl transform of \mathcal{H} , which is free of interband terms, is the equation defining the magnetic Wannier function on the corresponding equation defining the magnetic Bloch function. In the magnetic Wannier function representation we have

$$\mathcal{H} = \sum_{\lambda, \vec{q}', z, B} \left[\left\langle \lambda, \vec{q}', z, B \right| \mathcal{H}_0 + \Sigma(z) \left| \lambda, \vec{q}, z, B \right\rangle \right] \left| \lambda, \vec{q}', z, B \right\rangle \left\langle \lambda, \vec{q}, z, B \right| \quad (104)$$

where the matrix elements of $\mathcal{H}_0 + \Sigma(z)$ in general have dual magnetic Wannier functions on the left- and right-hand sides instead of complex conjugate of the same wave function. The equation defining $|\lambda, \vec{q}, z, B\rangle$ becomes

$$\mathcal{H} \left| \lambda, \vec{q}, z, B \right\rangle = \sum_{\vec{q}'} \left[\left\langle \lambda, \vec{q}', z, B \right| \mathcal{H}_0 + \Sigma(z) \left| \lambda, \vec{q}, z, B \right\rangle \right] \left| \lambda, \vec{q}', z, B \right\rangle \quad (105)$$

and that for $\langle \lambda, \vec{q}', z, B |$ is

$$\langle \lambda, \vec{q}', z, B | \mathcal{H} = \sum_{\vec{q}} \left[\left\langle \lambda, \vec{q}', z, B \right| \mathcal{H}_0 + \Sigma(z) \left| \lambda, \vec{q}, z, B \right\rangle \right] \langle \lambda, \vec{q}, z, B | \quad (106)$$

The form of the matrix elements is given in Appendix B of Ref.⁶², which may be written as

$$\langle \lambda, \vec{q}', z, B | \mathcal{H}_0 + \Sigma(z) \left| \lambda, \vec{q}, z, B \right\rangle = \exp \left[\left(\frac{ie}{\hbar c} \right) \vec{A}(\vec{q}) \cdot \vec{q} \right] H_\lambda(\vec{q} - \vec{q}', B, z) \delta_{\lambda\lambda'} \quad (107)$$

The magnetic translation operator is define by

$$\left| \lambda, \vec{q}, z, B \right\rangle = T(-\vec{q}) \left| \lambda, 0, z, B \right\rangle_0 = \exp \left[\left(\frac{-ie}{\hbar c} \right) \vec{A}(\vec{r}) \cdot \vec{q} \right] \left| \lambda, -\vec{q}, z, B \right\rangle_0 \quad (108)$$

where $T(-\vec{q})$ is the magnetic translation operator and $w_\lambda(\vec{r} - \vec{q}, z, B) \equiv \left| \lambda, -\vec{q}, z, B \right\rangle_0$ is the modified Wannier function centered at the lattice point \vec{q} . The equation satisfied by $w_\lambda(\vec{r} - 0, z, B) \equiv w_\lambda(\vec{r}, z, B)$ can explicitly be written

$$\mathcal{H}_0 w_\lambda(\vec{r}, z, B) + \int d^3r' \Sigma(\vec{r}, \vec{r}', z, B) w_\lambda(\vec{r}', z, B) = \sum_{\vec{q}'} H_\lambda(-\vec{q}', z, B) \left| \lambda, \vec{q}', z, B \right\rangle \quad (109)$$

and hence,

$$\mathcal{H} \left| \lambda, \vec{q}, z, B \right\rangle = T(-q) \mathcal{H} \left| \lambda, 0, z, B \right\rangle \quad (110)$$

which can be written explicitly as

$$\begin{aligned}
& \exp \left[\left(\frac{-ie}{\hbar c} \right) \vec{A}(\vec{r}) \cdot \vec{q} \right] \mathcal{H}_0(\vec{r} - \vec{q}) w_\lambda(\vec{r} - \vec{q}, z, B) \\
& + \exp \left[\left(\frac{-ie}{\hbar c} \right) \vec{A}(\vec{r}) \cdot \vec{q} \right] \int d^3 r' \Sigma(\vec{r} - \vec{q}, \vec{r}', z, B) w_\lambda(\vec{r}' - 0, z, B) \\
& = \sum_{\vec{q}'} \exp \left[\left(\frac{ie}{\hbar c} \right) \vec{A}(\vec{q}) \cdot \vec{q}' \right] H_\lambda(\vec{q} - \vec{q}', z, B) \exp \left[\left(\frac{-ie}{\hbar c} \right) \vec{A}(\vec{r}) \cdot \vec{q}' \right] w_\lambda(\vec{r} - \vec{q}', z, B) \quad (111)
\end{aligned}$$

Changing the variable of integration \vec{r}' to $\vec{r}' - \vec{q}$, noting that by symmetry,

$$\Sigma(\vec{r}, \vec{r}', z, B) = \exp \left[\left(\frac{-ie}{\hbar c} \right) \vec{A}(\vec{r}) \cdot \vec{r}' \right] \tilde{\Sigma}(\vec{r}, \vec{r}', z, B) \quad (112)$$

where $\Sigma(\vec{r} - \vec{q}, \vec{r}' - \vec{q}, z, B) = \Sigma(\vec{r}, \vec{r}', z, B)$. Dividing both sides of the equation by $\exp \left[\left(\frac{-ie}{\hbar c} \right) \vec{A}(\vec{r}) \cdot \vec{q} \right]$ and taking the lattice Fourier transform [i.e., multiply both sides by operation, $\left(\frac{1}{N\hbar} \right)^3 \sum_{\vec{q}} \exp \left(\frac{i}{\hbar} \vec{p} \cdot \vec{q} \right)$], we obtain

$$\begin{aligned}
& \mathcal{H}_0 \left(\vec{p} - \frac{e}{c} \vec{A}(\vec{r} + i\nabla_{\vec{k}}), \vec{r} \right) b_\lambda(\vec{r}, \vec{k}, B, z) \\
& + \int d^3 r' \exp \left[\left(\frac{-ie}{\hbar c} \right) \vec{A}(\vec{r}) \cdot \vec{r}' \right] \tilde{\Sigma}(\vec{r}, \vec{r}', z, B) b_\lambda(\vec{r}', \vec{k} + \frac{e}{\hbar c} \vec{A}(\vec{r} - \vec{r}'), B, z) \\
& = \sum_{\vec{q}} e^{i\vec{k} \cdot \vec{q}} H_\lambda(\vec{q}, z, B) e^{\left(\frac{ie}{\hbar c} \right) \vec{A}(\vec{r}) \cdot \vec{q}} b_\lambda \left(\vec{r}', \vec{k} + \frac{e}{\hbar c} \vec{A}(\vec{q}); B, z \right) \quad (113)
\end{aligned}$$

where the modified Bloch function $b_\lambda(\vec{r}', \vec{k} + \frac{e}{\hbar c} \vec{A}(\vec{q}); B, z)$ is defined by

$$\begin{aligned}
b_\lambda(\vec{r}, \vec{k}, B, z) &= (N\hbar^3)^{-\frac{1}{2}} \sum_{\vec{q}} e^{i\vec{k} \cdot \vec{q}} w_\lambda(\vec{r} - \vec{q}, z, B) \\
&\equiv e^{i\vec{k} \cdot \vec{r}} u_\lambda(\vec{r}, \vec{k}, z, B) \quad (114)
\end{aligned}$$

The equation satisfied by $u_\lambda(\vec{r}, \vec{k}, z, B)$ is

$$\begin{aligned}
& \mathcal{H}_0 \left(\vec{p} + \hbar \vec{k} - \frac{e}{c} \vec{A}(i\nabla_{\vec{k}}); \vec{r} \right) u_\lambda(\vec{r}, \vec{k}, z, B) \\
& + \int d^3 r' e^{i\vec{k} \cdot (\vec{r}' - \vec{r})} \tilde{\Sigma}(\vec{r}, \vec{r}', z, B) u_\lambda \left(\vec{r}, \vec{k} + \frac{e}{\hbar c} \vec{A}(\vec{r} - \vec{r}'), z, B \right) \\
& = \sum_{\vec{q}} e^{i\vec{k} \cdot \vec{q}} H_\lambda(\vec{q}, z, B) u_\lambda \left(\vec{r}, \vec{k} + \frac{e}{\hbar c} \vec{A}(\vec{q}), z, B \right) \quad (115)
\end{aligned}$$

F. Expansions in Powers of B

Let us expand $\mathcal{H}_0, u_\lambda(\vec{r}, \vec{k}, z, B), \tilde{\Sigma}(\vec{r}, \vec{r}', z, B)$ in powers of the magnetic field, B , as well

$$u_\lambda \left(\vec{r}, \vec{k} + \frac{e}{\hbar c} \vec{A}(\vec{q}), z, B \right) = \exp \left[\left(\frac{e}{\hbar c} \right) \vec{A}(\vec{q}) \cdot \nabla_{\vec{k}} \right] u_\lambda(\vec{r}, \vec{k}, z, B).$$

We expand both sides of Eq. (115) up to second order in B and equate the coefficients on both sides. Multiplying both sides of the resulting equations by the dual set of wave functions $\langle u_\lambda(\vec{r}, \vec{k}, z, B) |$, determined by Eq. (72) biorthogonal to $|u_\lambda(\vec{r}, \vec{k}, z, B)\rangle$, and integrating, we obtain for $\delta = \lambda$ the expression for $H_\lambda^{(1)}(k, z)$ and $H_\lambda^{(2)}(k, z)$, where

$$H_\lambda^{(l)}(\vec{k}, z) = \sum_{\vec{q}} e^{i\vec{k} \cdot \vec{q}} H_\lambda^{(l)}(\vec{q}, z) \quad (116)$$

We obtained the following expressions:

$$\begin{aligned} H_\lambda^{(1)}(k, z) = & \left\langle u_\lambda^0 \left| \mathcal{H}_0^{(1)} + \frac{e}{B\hbar c} \left[\vec{A}(\nabla_{\vec{k}}) \tilde{H}_\lambda^0(\vec{k}, z) \right] \cdot \nabla_{\vec{k}} \right| u_\lambda^0 \right\rangle \\ & + \left\langle u_\lambda^0(\vec{r}, \vec{k}, z) \left| \int d^3 r' e^{i\vec{k} \cdot (\vec{r}' - \vec{r})} \tilde{\Sigma}^0(\vec{r}, \vec{r}', z) \frac{e}{B\hbar c} \vec{A}(\vec{r} - \vec{r}') \cdot \nabla_{\vec{k}} \right| u_\lambda^0(\vec{r}', \vec{k}, z) \right\rangle \\ & + \left\langle u_\lambda^0(\vec{r}, \vec{k}, z) \left| \int d^3 r' e^{i\vec{k} \cdot (\vec{r}' - \vec{r})} \tilde{\Sigma}^{(1)}(\vec{r}, \vec{r}', z) \right| u_\lambda^0(\vec{r}', \vec{k}, z) \right\rangle \end{aligned} \quad (117)$$

$$\begin{aligned} H_\lambda^{(2)}(k, z) = & \left\langle u_\lambda^0 \left| \mathcal{H}_0^{(2)} \right| u_\lambda^0 \right\rangle + \left\langle u_\lambda^0 \left| \mathcal{H}_0^{(1)} \right| u_\lambda^{(1)} \right\rangle \\ & + \left\langle u_\lambda^0 \left| \sum_{\vec{q}} e^{i\vec{k} \cdot \vec{q}} \tilde{\mathcal{H}}_\lambda^0(\vec{q}, z) \frac{1}{2!} \left(\frac{e}{B\hbar c} \vec{A}(\vec{q}) \cdot \nabla_{\vec{k}} \right)^2 \right| u_\lambda^0 \right\rangle \\ & + \left\langle u_\lambda^0 \left| \frac{e}{B\hbar c} \vec{A}(\vec{q}) \cdot \nabla_{\vec{k}} \tilde{\mathcal{H}}_\lambda^0(\vec{q}, z) \left(\frac{e}{B\hbar c} \vec{A}(\vec{q}) \cdot i\nabla_{\vec{k}} \right) \right| u_\lambda^{(1)} \right\rangle \\ & + \left\langle u_\lambda^0 \left| \frac{e}{B\hbar c} \vec{A}(\vec{q}) \cdot \nabla_{\vec{k}} \mathcal{H}_\lambda^{(1)}(\vec{q}, z) \left(\frac{e}{B\hbar c} \vec{A}(\vec{q}) \cdot i\nabla_{\vec{k}} \right) \right| u_\lambda^0 \right\rangle \\ & - \mathcal{H}_\lambda^{(1)}(\vec{q}, z) \left\langle u_\lambda^0 \left| u_\lambda^{(1)} \right\rangle \right. \\ & + \left\langle u_\lambda^0(\vec{r}, \vec{k}, z) \left| \int d^3 r' e^{i\vec{k} \cdot (\vec{r}' - \vec{r})} \tilde{\Sigma}^0(\vec{r}, \vec{r}', z) \frac{1}{2!} \left(\frac{e}{B\hbar c} \vec{A}(\vec{r} - \vec{r}') \cdot \nabla_{\vec{k}} \right)^2 \right| u_\lambda^0(\vec{r}, \vec{k}, z) \right\rangle \\ & + \left\langle u_\lambda^0(\vec{r}, \vec{k}, z) \left| \int d^3 r' e^{i\vec{k} \cdot (\vec{r}' - \vec{r})} \tilde{\Sigma}^0(\vec{r}, \vec{r}', z) \left(\frac{e}{B\hbar c} \vec{A}(\vec{r} - \vec{r}') \cdot \nabla_{\vec{k}} \right) \right| u_\lambda^1(\vec{r}, \vec{k}, z) \right\rangle \\ & + \left\langle u_\lambda^0(\vec{r}, \vec{k}, z) \left| \int d^3 r' e^{i\vec{k} \cdot (\vec{r}' - \vec{r})} \tilde{\Sigma}^1(\vec{r}, \vec{r}', z) \left(\frac{e}{B\hbar c} \vec{A}(\vec{r} - \vec{r}') \cdot \nabla_{\vec{k}} \right) \right| u_\lambda^0(\vec{r}, \vec{k}, z) \right\rangle \\ & + \left\langle u_\lambda^0(\vec{r}, \vec{k}, z) \left| \int d^3 r' e^{i\vec{k} \cdot (\vec{r}' - \vec{r})} \tilde{\Sigma}^1(\vec{r}, \vec{r}', z) \right| u_\lambda^1(\vec{r}, \vec{k}, z) \right\rangle \\ & + \left\langle u_\lambda^0(\vec{r}, \vec{k}, z) \left| \int d^3 r' e^{i\vec{k} \cdot (\vec{r}' - \vec{r})} \tilde{\Sigma}^2(\vec{r}, \vec{r}', z) \right| u_\lambda^0(\vec{r}, \vec{k}, z) \right\rangle \end{aligned} \quad (118)$$

Note that in the expression for $H_\lambda^{(2)}(k, z)$ we need $|u_\lambda^{(1)}(\vec{r}, \vec{k}, z)\rangle$, which can be written

$$|u_\lambda^1(\vec{r}, \vec{k}, z)\rangle = \sum_{\sigma} \beta_{\lambda\sigma} |u_\sigma^0(\vec{r}, \vec{k}, z)\rangle \quad (119)$$

and for $\lambda \neq \delta$, $\beta_{\lambda\sigma}$ can be determined from the same set of equations which determined $H_\lambda^{(i)}(k, z)$. It is given by

$$\begin{aligned} \langle u_\sigma^0(\vec{r}, \vec{k}, z) | u_\lambda^{(1)}(\vec{r}, \vec{k}, z) \rangle &= -[H_\sigma^0(k, z) - H_\lambda^0(k, z)]^{-1} \langle u_\sigma^0(\vec{r}, \vec{k}, z) | H_\lambda^{(1)op} | u_\lambda^0(\vec{r}, \vec{k}, z) \rangle \\ &= \beta_{\sigma\lambda} \end{aligned} \quad (120)$$

where $\langle u_\sigma^0 | H_\lambda^{(1)op} | u_\lambda^0 \rangle$ is given by Eq. (117) with the band index λ replaced by δ on the left-hand wave function. For $\lambda = \delta$, $\beta_{\lambda\lambda}$ can be obtained from the requirement that the magnetic Wannier functions are biorthogonal. This is expressed by the equation

$$\int d^3r \exp \left[\left(\frac{1}{2} \frac{ie}{\hbar c} \right) (\vec{B} \times \vec{r}) \cdot (\vec{q}' - \vec{q}) \right] \Omega_\lambda^*(\vec{r} - \vec{q}', z, B) w_\lambda(\vec{r} - \vec{q}, z, B) = \delta_{\vec{q} \vec{q}'}, \quad (121)$$

where we have written

$$\langle \lambda, \vec{q}', z, B | = \exp \left[\left(\frac{1}{2} \frac{ie}{\hbar c} \right) (\vec{B} \times \vec{r}) \cdot \vec{q}' \right] \Omega_\lambda^*(\vec{r} - \vec{q}', z, B) \quad (122)$$

Expanding the left-hand side of Eq. (121) in powers of B we obtain the following relations:

$$\langle \lambda, \vec{q}', z | \lambda, \vec{q}, z \rangle = \delta_{\vec{q} \vec{q}'}, \quad (123)$$

$$\begin{aligned} \langle \lambda, \vec{q}', z | w_\lambda^{(1)}(\vec{r} - \vec{q}', z) \rangle + \langle \Omega_\lambda^{(1)*}(\vec{r} - \vec{q}', z) | \lambda, \vec{q}, z \rangle \\ + \frac{1}{2} \left(\frac{ie}{\hbar c} \right) \langle (\hat{z} \times \vec{r}) \cdot (\vec{q}' - \vec{q}) \Omega_\lambda^{0*}(\vec{r} - \vec{q}', z) w_\lambda^0(\vec{r} - \vec{q}, z) \rangle = 0 \end{aligned} \quad (124)$$

where $\Omega_\lambda^{0*}(\vec{r} - \vec{q}', z) \implies \langle \lambda, \vec{q}, z |$ and $w_\lambda^0(\vec{r} - \vec{q}, z) \implies | \lambda, \vec{q}, z \rangle$.

G. Berry connection and Berry curvature

By virtue of the identity

$$(\hat{z} \times \vec{r}) \cdot (\vec{q}' - \vec{q}) = \hat{z} \cdot [(\vec{r} - \vec{q}') \times (\vec{r} - \vec{q}) - (\vec{q}' \times \vec{q})] \quad (125)$$

and of Eq. (123), we obtain from Eq. (124) after lattice Fourier transformation,

$$\langle u_\lambda^0 | u_\lambda^{(1)} \rangle + \langle u_\lambda^{(1)} | u_\lambda^0 \rangle = \frac{1}{2} \left(\frac{e}{\hbar c} \right) \left(\frac{\partial}{\partial \vec{k}_y} X_\lambda - \frac{\partial}{\partial \vec{k}_x} Y_\lambda \right) \quad (126)$$

where

$$X_\lambda = \left\langle u_\lambda^0 \left| i \frac{\partial}{\partial \vec{k}_x} u_\lambda^0 \right. \right\rangle \quad (127)$$

$$Y_\lambda = \left\langle u_\lambda^0 \left| i \frac{\partial}{\partial \vec{k}_y} u_\lambda^0 \right. \right\rangle \quad (128)$$

which resemble the *Berry connections* in modern terminology, and thus the terms in the parenthesis of Eq. (126) resembles *Berry curvature*. Indeed, the Berry curvature of Bloch states physically represents part of their perturbative response to uniform electromagnetic fields. Equation (126) yields the expression for $\beta_{\lambda\lambda}$,

$$\beta_{\lambda\lambda} = \frac{1}{4} \left(\frac{e}{\hbar c} \right) \left(\frac{\partial}{\partial \vec{k}_y} X_\lambda - \frac{\partial}{\partial \vec{k}_x} Y_\lambda \right) \quad (129)$$

Thus, $H_\lambda^{(1)}(k, z)$ and $H_\lambda^{(2)}(k, z)$ are completely determined in the expansion of $\tilde{H}_\lambda \left(\vec{p} - \frac{e}{c} \vec{A}(\vec{q}); B, z \right)$ in Eq. (78). The lattice Weyl transform, which is free of interband terms, is obtained by replacement of $\hbar \vec{k}$ by $\left[\hbar \vec{k} - \frac{e}{c} \vec{A}(\vec{q}) \right]$ in $H_\lambda^{(1)}(k, z)$ and $H_\lambda^{(2)}(k, z)$. Equation (78) written up to second order in its explicit dependence in B , beyond the vector potential, is thus given by

$$\begin{aligned} \tilde{H}_\lambda \left(\vec{p} - \frac{e}{c} \vec{A}(\vec{q}), B, z \right) &= \tilde{H}_\lambda^0 \left(\vec{p} - \frac{e}{c} \vec{A}(\vec{q}), z \right) + B H_\lambda^{(1)} \left(\vec{p} - \frac{e}{c} \vec{A}(\vec{q}), z \right) \\ &+ B^2 H_\lambda^{(2)} \left(\vec{p} - \frac{e}{c} \vec{A}(\vec{q}), z \right) + \dots \end{aligned} \quad (130)$$

H. Derivation of General Expression for Many-Body χ

In this section, will derive the most general expression for χ using the temperature Green's function formalism of Luttinger and Ward.⁶³ The magnetic susceptibility for a system of volume V is,

$$\chi = \frac{1}{V} \lim_{B \Rightarrow 0} \frac{\partial^2}{\partial B^2} \left(\frac{1}{\beta} \ln Z \right) \quad (131)$$

which at zero temperature can be expressed as,

$$\chi = \frac{1}{V} \lim_{\substack{B \Rightarrow 0 \\ \beta \Rightarrow \infty}} \frac{\partial^2}{\partial B^2} \left(\frac{\partial}{\partial \beta} \ln Z \right) \quad (132)$$

The expression for $\ln Z$ as a functional of the temperature Green's function, \mathcal{G}_{ζ_l} , is given by Luttinger and Ward,⁶³

$$-\ln Z = \Phi(\mathcal{G}_{\zeta_l}) - Tr \Sigma(\mathcal{G}_{\zeta_l}) \mathcal{G}_{\zeta_l} + Tr \ln(-\mathcal{G}_{\zeta_l}) \quad (133)$$

where the temperature Green's-function operator $\mathcal{G}_{\zeta_l}^{-1}$ is defined formally by

$$\mathcal{G}_{\zeta_l}^{-1} = \zeta_l - \mathcal{H}_0 - \Sigma_{\zeta_l}, \quad (134)$$

$$\zeta_l = (2l+1) \frac{\pi i}{\beta} + \mu. \quad (135)$$

Tr is defined as $\sum_l \tilde{T}r$, where $\tilde{T}r$ refers to taking the trace in any convenient representation. The functional $\Phi(\mathcal{G}_{\zeta_l})$ is defined as

$$\Phi(\mathcal{G}_{\zeta_l}) = \lim_{\lambda \Rightarrow 1} Tr \sum_n \frac{\lambda^n}{2n} \Sigma^{(n)}(\mathcal{G}_{\zeta_l}) \mathcal{G}_{\zeta_l} \quad (136)$$

$\Sigma^{(n)}(\mathcal{G}_{\zeta_l})$ is the n th-order self-energy part where only the interaction parameter λ occurring explicitly in (136) is used to determine the order.

It is convenient to work in the coordinate representation as a first step to simplify the right-hand side of Eq. (131). The total Hamiltonian in this representation takes the form

$$\mathcal{H} = \int d^3r \psi_\alpha^\dagger(\vec{r}) \mathcal{H}_0 \psi_\alpha(\vec{r}) + \frac{1}{2} \int d^3r d^3r' \psi_\alpha^\dagger(\vec{r}) \psi_\beta^\dagger(\vec{r}') v_{\alpha\beta\gamma\delta}(\vec{r}, \vec{r}') \psi_\gamma(\vec{r}') \psi_\delta(\vec{r}) \quad (137)$$

where repeated spin indices are summed over, and for simplicity, we may take Eq. (55) for \mathcal{H}_0 . The term $v_{\alpha\beta\gamma\delta}(\vec{r}, \vec{r}')$ is the interaction between a pair of particles assumed to be velocity independent; this immediately implies that in coordinate representation the field dependence of $\ln Z$ in (133) occurs only through the field dependence of $\mathcal{G}_{\zeta_l}(r, r')$. To take spin into account explicitly, both $\mathcal{G}_{\zeta_l}(r, r')$ and Σ_{ζ_l} must be considered as 2×2 matrices in spin indices. The form of $\mathcal{G}_{\zeta_l}(r, r')$ and Σ_{ζ_l} is given by Eq. (112) by gauge invariance. It is convenient for our purpose to expand Σ_{ζ_l} in powers of its explicit dependence on the magnetic field (beyond the Peierls phase factor) and write

$$\Sigma_{\zeta_l}(\vec{r}, \vec{r}') = \Sigma_{\zeta_l}^0(\vec{r}, \vec{r}') + B \Sigma_{\zeta_l}^{(1)}(\vec{r}, \vec{r}') + B^2 \Sigma_{\zeta_l}^{(2)}(\vec{r}, \vec{r}') + \dots \quad (138)$$

where the remaining field dependence of $\Sigma_{\zeta_l}^0(\vec{r}, \vec{r}')$, $\Sigma_{\zeta_l}^{(1)}(\vec{r}, \vec{r}')$, and $\Sigma_{\zeta_l}^{(2)}(\vec{r}, \vec{r}')$ occurs only through the Peierls phase factor.

We have, using the definition of $\Phi(\mathcal{G}_{\zeta_l})$, the following relations:

$$\left. \frac{\partial^2 \Phi(\mathcal{G}_{\zeta_l})}{\partial B^2} \right|_{B \Rightarrow 0} = \sum_l \int d^3r d^3r' \left(\tilde{\Sigma}_{\zeta_l}^{(1)}(\vec{r}, \vec{r}') \frac{\partial \tilde{\mathcal{G}}_{\zeta_l}(\vec{r}, \vec{r}', B)}{\partial B} \right|_{B \Rightarrow 0} + \tilde{\Sigma}_{\zeta_l}^0(\vec{r}, \vec{r}') \frac{\partial^2 \tilde{\mathcal{G}}_{\zeta_l}(\vec{r}, \vec{r}', B)}{\partial B^2} \Big|_{B \Rightarrow 0} \right) \quad (139)$$

$$\begin{aligned} \left. \frac{\partial^2 Tr(\Sigma_{\zeta_l} \mathcal{G}_{\zeta_l})}{\partial B^2} \right|_{B \Rightarrow 0} = & - \sum_l \int d^3r d^3r' \left(\tilde{\Sigma}_{\zeta_l}^{(1)}(\vec{r}, \vec{r}') \tilde{\mathcal{G}}_{\zeta_l}(\vec{r}, \vec{r}', B) \Big|_{B \Rightarrow 0} + 2 \tilde{\Sigma}_{\zeta_l}^{(1)}(\vec{r}, \vec{r}') \frac{\partial \tilde{\mathcal{G}}_{\zeta_l}(\vec{r}, \vec{r}', B)}{\partial B} \Big|_{B \Rightarrow 0} \right. \\ & \left. + \tilde{\Sigma}_{\zeta_l}^0(\vec{r}, \vec{r}') \frac{\partial^2 \tilde{\mathcal{G}}_{\zeta_l}(\vec{r}, \vec{r}', B)}{\partial B^2} \Big|_{B \Rightarrow 0} \right) \end{aligned} \quad (140)$$

where $\tilde{\Sigma}_{\zeta_l}^{(i)}(\vec{r}, \vec{r}')$ and $\tilde{\Sigma}_{\zeta_l}^0(\vec{r}, \vec{r}')$ are field-independent quantities. The above relations lead to a more convenient expression for χ ,

$$\chi = -\frac{1}{V} \left(\frac{\partial^2}{\partial B^2} \frac{1}{\beta} Tr \ln(-\mathcal{G}_{\zeta_l}) \right) \Big|_{B \Rightarrow 0} + \frac{1}{V} \left(\frac{1}{\beta} Tr 2 \tilde{\Sigma}_{\zeta_l}^{(2)} \tilde{\mathcal{G}}_{\zeta_l} + \frac{1}{\beta} Tr \tilde{\Sigma}_{\zeta_l}^{(1)} \frac{\partial \tilde{\mathcal{G}}_{\zeta_l}}{\partial B} \right) \Big|_{B \Rightarrow 0} \quad (141)$$

The first term in Eq. (141) has exactly the same form as that of the noninteracting Fermi systems, except for the replacement of the “noninteracting” \mathcal{G}_{ζ_l} by the exact \mathcal{G}_{ζ_l} for the interacting, free, or Bloch, electrons. The second term can be immediately recognized as correction to the “crystalline induced diamagnetism” as calculated by the first term. The last term turns out to contain corrections to both the “crystalline paramagnetism” and “crystalline induced diamagnetism” as calculated from the first term in Eq. (141).

We are, in the present case, of course, interested in the effective one-particle Schrodinger Hamiltonian

$$\mathcal{H}_{\zeta_l} = \mathcal{H}_0 + \Sigma_{\zeta_l} \quad (142)$$

which is formally the same as that of Eq. (54), with the replacement $z \Rightarrow \zeta_l$ (we have chosen to indicate the discrete frequency dependence of operators by a subscript). Therefore, all the results of Sec. VIB can be formally carried over to apply to the effective Hamiltonian given above. The beauty and power in the use of the Weyl transform is that the Weyl transform of an operator is a physically meaningful quantity and faithfully corresponds to the original quantum-mechanical operator. Moreover, it provides a natural way of calculating the trace of any function of \mathcal{H}_{ζ_l} , as a power-series expansion in \hbar , Planck’s constant, which is equivalent to an expansion in the magnetic field strength for Fermi systems possessing translational symmetry.

$$\begin{aligned} \tilde{Tr} F(\mathcal{H}_{\zeta_l}) = & \left(\frac{1}{2\pi} \right)^3 \sum_{\lambda} \int d^3k \, d^3q \left\{ F\left(\tilde{H}_{\lambda}(k, B, \zeta_l) \right) \right. \\ & \left. - \frac{1}{24} \left(\frac{eB}{\hbar c} \right)^2 F''(\tilde{H}_{\lambda}^0(k, \zeta_l)) \left[\frac{\partial^2 \tilde{H}_{\lambda}^0}{\partial \vec{k}_x^2} \frac{\partial^2 \tilde{H}_{\lambda}^0}{\partial \vec{k}_y^2} - \left(\frac{\partial^2 \tilde{H}_{\lambda}^0}{\partial \vec{k}_x \partial \vec{k}_y} \right)^2 \right] \right\} + O(B^4) \quad (143) \end{aligned}$$

where, $\vec{p} - (e/c)A(\vec{q})$ in (130) is replaced by $\hbar\vec{k}$ in Eq. (143). In the above expression, it is assumed that $\tilde{H}_{\lambda}^0(k, \zeta_l)$ is diagonal in spin indices; this is generally true for nonferromagnetic systems. The first term in Eq. (143) can then be expanded up to second order in B using the expansion given in Eq. (130). Applying this result to the first term of Eq. (141), we obtain

$$- \frac{1}{V} \left(\frac{\partial^2}{\partial B^2} \frac{1}{\beta} Tr \ln(-\mathcal{G}_{\zeta_l}) \right)_{B \Rightarrow 0} = \chi_{LP} + \chi_{CP} + \chi_{ID} \quad (144)$$

where

$$\chi_{LP} = \frac{1}{12} \left(\frac{e}{\hbar c} \right)^2 \sum_{\lambda} \left(\frac{1}{2\pi} \right)^3 \int d^3k \, k_B T \sum_l \left[\frac{\partial^2 \tilde{H}_{\lambda}^0}{\partial \vec{k}_x^2} \frac{\partial^2 \tilde{H}_{\lambda}^0}{\partial \vec{k}_y^2} - \left(\frac{\partial^2 \tilde{H}_{\lambda}^0}{\partial \vec{k}_x \partial \vec{k}_y} \right)^2 \right] (\mathcal{G}_{\zeta_l}(k, \zeta_l))^2 \quad (145)$$

$$\chi_{CP} = - \sum_{\lambda} \left(\frac{1}{2\pi} \right)^3 \int d^3k \, k_B T \sum_l \left[\tilde{H}_{\lambda}^{(1)}(k, \zeta_l) \right]^2 \left(\mathcal{G}_{\zeta_l}(k, \zeta_l) \right)^2 \quad (146)$$

$$\chi_{ID} = - \sum_{\lambda} \left(\frac{1}{2\pi} \right)^3 \int d^3k \, k_B T \sum_l 2\tilde{H}_{\lambda}^{(2)}(k, \zeta_l) \mathcal{G}_{\zeta_l}(k, \zeta_l) \quad (147)$$

$$\mathcal{G}_{\zeta_l}(k, \zeta_l) = \left[\zeta_l - \tilde{H}_{\lambda}^0(k, \zeta_l) \right]^{-1} \quad (148)$$

In Eqs. (145), (146), and (147), taking the trace over spin indices is implied. χ_{LP} is a generalized Landau- Peierls formula for the orbital diamagnetism of free and Bloch electrons. It is for the case of an interacting free-electron gas that this term was derived by Fukuyama and McClure.⁶⁴ In the limit of vanishing self-energy parts, Eq. (144) exactly reproduces the expression for χ of Bloch electrons, both with or without spin-orbit coupling, as given by Roth,⁵⁴ and by Wannier and Upadhyaya,⁵⁵ respectively. Moreover, when the self-energy part is assumed to be independent of ζ_l , which holds true in Hartree-Fock approximation, the form of Eq. (144), after summation over ζ_l , is exactly the same as that of the noninteracting case.

χ_{CP} , which includes the effect of free-electron spin and g -factor, will be referred to as the crystalline paramagnetism, and χ_{ID} , the induced diamagnetism, although its sign cannot be determined *a priori* even in the Hartree-Fock approximation and in the noninteracting case.

We consider the correction terms represented by the last two terms of Eq. (141) explicitly from the self-energies, $\tilde{\Sigma}_{\zeta_l}^{(i)}(\vec{r}, \vec{r}')$. As we have mentioned earlier, these corrections only modify χ_{CP} and χ_{ID} but do not affect χ_{LP} . Let us recast the last two terms of Eq. (141), which we now denote by χ_{cor} and write them as follows:

$$\chi_{cor} = \frac{1}{V} \lim_{B \Rightarrow 0} \frac{\partial}{\partial B} Tr \frac{1}{\beta} \left(\tilde{\Sigma}_{\zeta_l}^{(1)} + 2B \tilde{\Sigma}_{\zeta_l}^{(2)} \right) \mathcal{G}_{\zeta_l} \quad (149)$$

Recall that in the coordinate representation, the Peierls phase factors occurring in $\tilde{\Sigma}_{\zeta_l}^{(i)}$ and \mathcal{G}_{ζ_l} cancel. However, it is convenient to retain these phase factors in Eq. (149) as the trace will now be taken using the biorthogonal magnetic function representation discussed in Sec. VIB. The trace would then be expressed in terms of the Weyl transform, where indeed the Weyl transform of \mathcal{G}_{ζ_l} is diagonal in band indices, resulting in much simplification. We have

$$\chi_{cor} = \frac{1}{V} \lim_{B \Rightarrow 0} \frac{\partial}{\partial B} \sum_{\lambda} \left(\frac{1}{2\pi} \right)^3 \int d^3k \, k_B T \sum_l \left[\left(\tilde{\Sigma}_{\lambda\lambda}^{(1)}(\vec{k}, B, \zeta_l) + 2B \tilde{\Sigma}_{\lambda\lambda}^{(2)}(\vec{k}, B, \zeta_l) \right) \right] \mathcal{G}_{\lambda}(\vec{k}, B, \zeta_l) \quad (150)$$

where in the last equation a familiar change of variable has been made, $p - (e/c)A(q) \implies \hbar k$, and from Eq. (86) we have

$$\mathcal{G}_\lambda(\vec{k}, B, \zeta_l) = \left[\zeta_l - \tilde{H}_\lambda(\vec{k}, B, \zeta_l) \right]^{-1} + O(B^2) \quad (151)$$

$\tilde{H}_\lambda(\vec{k}, B, \zeta_l)$ is of the form given by Eq. (130) with the replacement $z \implies \zeta_l$. Since we need $\tilde{\Sigma}_{\lambda\lambda}^{(2)}(\vec{k}, B, \zeta_l)$ only to zero order in the field, the calculation of the second term in Eq. (150) is trivial. Denoting this contribution as $\chi_{cor}^{(2)}$ we have

$$\chi_{cor}^{(2)} = \sum_\lambda \left(\frac{1}{2\pi} \right)^3 \int d^3k \, k_B T \sum_l 2 \left\langle u_\lambda^0(\vec{r}, \vec{k}, \zeta_l) \left| \int d^3r' e^{i\vec{k} \cdot (\vec{r}' - \vec{r})} \tilde{\Sigma}_{\zeta_l}^{(2)}(\vec{r}, \vec{r}') u_\lambda^0(\vec{r}', \vec{k}, \zeta_l) \right\rangle \mathcal{G}_\lambda(\vec{k}, \zeta_l) \quad (152)$$

$\chi_{cor}^{(2)}$ is indeed a correction to χ_{ID} as can be seen from Eqs. (147) and (118).

To find $\tilde{\Sigma}_{\lambda\lambda}^{(1)}(\vec{k}, B, \zeta_l)$, we write down the effect of operating $\tilde{\Sigma}_{\zeta_l}^{(1)}$ on the magnetic Bloch function⁶² (same relation holds for $\tilde{\Sigma}_{\zeta_l}^{(2)}$)

$$\Sigma_{\zeta_l}^{(1)} \left| \vec{p}, \lambda, \zeta_l, B \right\rangle = \sum_{\vec{q}, \lambda'} e^{\frac{i\vec{p} \cdot \vec{q}}{\hbar}} \left[\Sigma_{\zeta_l}^{(1)}(\vec{q}, B) \right]_{\lambda\lambda'} \left| \vec{p} + \frac{e}{c} \vec{A}(\vec{q}), \lambda', \zeta_l, B \right\rangle \quad (153)$$

The Weyl transform of $\Sigma_{\zeta_l}^{(1)}$ is

$$\Sigma_{\lambda\lambda'}^{(1)}(\vec{p}, \vec{q}, \zeta_l, B) = \sum_{\vec{u}} e^{\frac{2i\vec{q} \cdot \vec{u}}{\hbar}} \left\langle \vec{p} + \vec{u}, \lambda, \zeta_l, B \left| \Sigma_{\zeta_l}^{(1)} \right| \vec{p} - \vec{u}, \lambda', \zeta_l, B \right\rangle \quad (154)$$

and by virtue of Eq. (153) we obtain

$$\begin{aligned} \Sigma_{\lambda\lambda'}^{(1)}(\vec{p}, \vec{q}, \zeta_l, B) &= \sum_{\vec{u}} e^{\frac{2i\vec{q} \cdot \vec{u}}{\hbar}} \sum_{\vec{v}} \delta_{\vec{u}, \frac{e}{2c} \vec{A}(\vec{v})} e^{\frac{i\vec{p} \cdot \vec{v}}{\hbar}} \left[\Sigma_{\lambda\lambda'}^{(1)}(\vec{v}, \zeta_l, B) \right] \\ &= \sum_{\vec{v}} \exp \left[\frac{1}{\hbar} \left(\vec{p} - \frac{e}{c} \vec{A}(\vec{q}) \right) \cdot \vec{v} \right] \Sigma_{\lambda\lambda'}^{(1)}(\vec{v}, \zeta_l, B) \end{aligned} \quad (155)$$

which in turn yields

$$\Sigma_{\lambda\lambda'}^{(1)}(\vec{p}, \vec{q}, \zeta_l, B) \Big|_{\vec{p} - \frac{e}{c} \vec{A}(\vec{q}) \implies \hbar \vec{k}} = \sum_{\vec{v}} \exp \left[\frac{1}{\hbar} \vec{k} \cdot \vec{v} \right] \Sigma_{\lambda\lambda'}^{(1)}(\vec{v}, \zeta_l, B) \quad (156)$$

We are therefore interested in the right-hand side of Eq. (156), to obtain this we may proceed in a manner quite similar to that used in Sec. VIB, i.e., Eqs. (109)-(118). However, at this stage, Eq. (153) provides a very good starting point. The relation between $|p, \lambda, \zeta_l, B\rangle$ and the modified Bloch function $b_\lambda(\vec{r}, \vec{k}, B, \zeta_l)$, used in the perturbation theory of Sec. VIB, can be easily deduced from Eqs. (108) and (114)

$$|p, \lambda, \zeta_l, B\rangle = b_\lambda \left(\vec{r}, \vec{k} - \frac{e}{\hbar c} \vec{A}(\vec{r}), \zeta_l, B \right) \quad (157)$$

Let us make the substitution $\vec{p} \Rightarrow \vec{p}' + (e/c)\vec{A}(\vec{r})$ in Eq. (153) and obtain the relation

$$\begin{aligned} & \int d^3r' \exp \left[\left(\frac{-ie}{\hbar c} \right) \vec{A}(\vec{r}) \cdot \vec{r}' \right] \tilde{\Sigma}_{\zeta_l}^{(1)}(\vec{r}, \vec{r}') b_{\lambda} \left(\vec{r}', \vec{p}' + \frac{e}{c} \vec{A}(\vec{r} - \vec{r}'), \zeta_l, B \right) \\ &= \sum_{\vec{q}, \lambda'} \exp \left[\left(\frac{i}{\hbar c} \right) \left(\vec{p}' + \frac{e}{c} \vec{A}(\vec{r}) \right) \cdot \vec{q} \right] \tilde{\Sigma}_{\lambda \lambda'}^{(1)}(\vec{q}, \zeta_l, B) b_{\lambda'} \left(\vec{r}, \vec{p}' + \frac{e}{c} \vec{A}(\vec{q}), \zeta_l, B \right) \end{aligned} \quad (158)$$

The equation in terms of the modified periodic function $u_{\lambda'}(\vec{r}, \vec{k}, \zeta_l, B)$ is therefore given by

$$\begin{aligned} & \int d^3r' \exp \left[i\vec{k} \cdot (\vec{r} - \vec{r}') \right] \tilde{\Sigma}_{\zeta_l}^{(1)}(\vec{r}, \vec{r}') u_{\lambda} \left(\vec{r}', \vec{k} + \frac{e}{\hbar c} \vec{A}(\vec{r} - \vec{r}'), \zeta_l, B \right) \\ &= \sum_{\vec{q}, \lambda'} \exp \left[i\vec{k} \cdot \vec{q} \right] \tilde{\Sigma}_{\lambda \lambda'}^{(1)}(\vec{q}, \zeta_l, B) u_{\lambda'} \left(\vec{r}, \vec{k} + \frac{e}{\hbar c} \vec{A}(\vec{q}), \zeta_l, B \right) \end{aligned} \quad (159)$$

Equation (159) corresponds to Eq. (115) of Sec. VIB. Perturbative treatment can then be carried out, using the expansion of $\tilde{\Sigma}_{\lambda \lambda'}^{(1)}(\vec{q}, \zeta_l, B)$ and $u_{\lambda'}(\vec{r}, \vec{k}, \zeta_l, B)$ in powers of B , and solution is obtained up to first order in B for $\tilde{\Sigma}_{\lambda \lambda}^{(1)}(\vec{k}, \zeta_l, B)$. Writing

$$\tilde{\Sigma}_{\lambda \lambda}^{(1)}(\vec{k}, \zeta_l, B) = \tilde{\Sigma}_{\lambda \lambda}^{(1)0}(\vec{k}, \zeta_l) + B \tilde{\Sigma}_{\lambda \lambda}^{(1)(1)}(\vec{k}, \zeta_l) + \dots \quad (160)$$

we get, by equating the zero- and first-order coefficients of B , the following relations:

$$\int d^3r' \exp \left[i\vec{k} \cdot (\vec{r} - \vec{r}') \right] \tilde{\Sigma}_{\zeta_l}^{(1)}(\vec{r}, \vec{r}') u_{\lambda}^0(\vec{r}', \vec{k}, \zeta_l) = \sum_{\vec{q}, \lambda'} \exp \left[i\vec{k} \cdot \vec{q} \right] \tilde{\Sigma}_{\lambda \lambda'}^{(1)0}(\vec{q}) u_{\lambda'}^0(\vec{r}, \vec{k}, \zeta_l) \quad (161)$$

$$\begin{aligned} & \int d^3r' \exp \text{Big} [i\vec{k} \cdot (\vec{r}' - \vec{r})] \tilde{\Sigma}_{\zeta_l}^{(1)}(\vec{r}, \vec{r}') u_{\lambda}^{(1)}(\vec{r}', \vec{k}, \zeta_l) \\ &+ \int d^3r' \exp \left[i\vec{k} \cdot (\vec{r}' - \vec{r}) \right] \tilde{\Sigma}_{\zeta_l}^{(1)}(\vec{r}, \vec{r}') \frac{e}{B\hbar c} A(\vec{r} - \vec{r}') \cdot \nabla_{\vec{k}} u_{\lambda}^0(\vec{r}', \vec{k}, \zeta_l) \\ &= \sum_{\vec{q}, \lambda'} \exp \left[i\vec{k} \cdot \vec{q} \right] \left(\tilde{\Sigma}_{\lambda \lambda'}^{(1)0}(\vec{q}, \zeta_l) \frac{e}{B\hbar c} A(\vec{q}) \cdot \nabla_{\vec{k}} u_{\lambda'}^0(\vec{r}, \vec{k}, \zeta_l) \right. \\ &\quad \left. + \tilde{\Sigma}_{\lambda \lambda'}^{(1)0}(\vec{q}, \zeta_l) u_{\lambda'}^{(1)}(\vec{r}', \vec{k}, \zeta_l) + \tilde{\Sigma}_{\lambda \lambda'}^{(1)(1)}(\vec{q}, \zeta_l) u_{\lambda'}^0(\vec{r}, \vec{k}, \zeta_l) \right) \end{aligned} \quad (162)$$

These relations yield for $\tilde{\Sigma}_{\lambda \lambda}^{(1)0}(\vec{q}, \zeta_l)$ and $\tilde{\Sigma}_{\lambda \lambda}^{(1)(1)}(\vec{q}, \zeta_l)$ the following expressions:

$$\tilde{\Sigma}_{\lambda \lambda}^{(1)0}(\vec{q}, \zeta_l) = \left\langle u_{\lambda}^0(\vec{r}, \vec{k}, \zeta_l) \left| \int d^3r' e^{i\vec{k} \cdot (\vec{r}' - \vec{r})} \tilde{\Sigma}_{\zeta_l}^{(1)}(\vec{r}, \vec{r}') u_{\lambda}^0(\vec{r}', \vec{k}, \zeta_l) \right. \right\rangle \quad (163)$$

$$\begin{aligned} \tilde{\Sigma}_{\lambda \lambda}^{(1)(1)}(\vec{q}, \zeta_l) &= \left\langle u_{\lambda}^0(\vec{r}, \vec{k}, \zeta_l) \left| \int d^3r' e^{i\vec{k} \cdot (\vec{r}' - \vec{r})} \tilde{\Sigma}_{\zeta_l}^{(1)}(\vec{r}, \vec{r}') u_{\lambda}^{(1)}(\vec{r}', \vec{k}, \zeta_l) \right. \right\rangle \\ &+ \left\langle u_{\lambda}^0(\vec{r}, \vec{k}, \zeta_l) \left| \int d^3r' e^{i\vec{k} \cdot (\vec{r}' - \vec{r})} \tilde{\Sigma}_{\zeta_l}^{(1)}(\vec{r}, \vec{r}') \frac{e}{B\hbar c} A(\vec{r} - \vec{r}') \cdot \nabla_{\vec{k}} u_{\lambda}^0(\vec{r}', \vec{k}, \zeta_l) \right. \right\rangle \\ &+ \left\langle u_{\lambda}^0(\vec{r}, \vec{k}, \zeta_l) \left| \sum_{\lambda'} \frac{e}{B\hbar c} A(\nabla_{\vec{k}}) \sum_{\vec{q}} e^{i\vec{k} \cdot \vec{q}} \tilde{\Sigma}_{\lambda \lambda'}^{(1)0}(\vec{q}, \zeta_l) \cdot i \nabla_{\vec{k}} u_{\lambda'}^0(\vec{r}', \vec{k}, \zeta_l) \right. \right\rangle \\ &- \sum_{\lambda'} \tilde{\Sigma}_{\lambda \lambda'}^{(1)0}(\vec{q}, \zeta_l) \left\langle u_{\lambda}^0(\vec{r}', \vec{k}, \zeta_l) \left| u_{\lambda'}^{(1)}(\vec{r}', \vec{k}, \zeta_l) \right. \right\rangle \end{aligned} \quad (164)$$

The first and last terms of Eq. (164) can be combined through the use of Eqs. (119), (120), and (161) to yield

$$\begin{aligned}
& \left\langle u_{\lambda}^0(\vec{r}, \vec{k}, \zeta_l) \left| \int d^3 r' e^{i\vec{k} \cdot (\vec{r}' - \vec{r})} \tilde{\Sigma}_{\zeta_l}^{(1)}(\vec{r}, \vec{r}') u_{\lambda}^{(1)}(\vec{r}', \vec{k}, \zeta_l) \right\rangle - \sum_{\lambda'} \Sigma_{\lambda\lambda'}^{(1)0}(\vec{k}, \zeta_l) \left\langle u_{\lambda}^0(\vec{r}', \vec{k}, \zeta_l) \left| u_{\lambda'}^{(1)}(\vec{r}', \vec{k}, \zeta_l) \right\rangle \right\rangle \\
&= - \sum_{\lambda' \neq \lambda} 2(\tilde{H}_{\lambda'}^0 - \tilde{H}_{\lambda}^0)^{-1} \langle u_{\lambda}^0 | \mathcal{H}_{\delta}^{(1)} | u_{\lambda'}^0 \rangle \langle u_{\lambda'}^0 | \mathcal{H}_{\delta}^{(1)} | u_{\lambda}^0 \rangle \\
&\quad - \sum_{\lambda' \neq \lambda} (\tilde{H}_{\lambda'}^0 - \tilde{H}_{\lambda}^0)^{-1} \left(\langle u_{\lambda'}^0 | \mathcal{H}_{\Delta}^{(1)} | u_{\lambda}^0 \rangle \langle u_{\lambda}^0 | \mathcal{H}_{\delta}^{(1)} | u_{\lambda'}^0 \rangle + \langle u_{\lambda}^0 | \mathcal{H}_{\Delta}^{(1)} | u_{\lambda'}^0 \rangle \langle u_{\lambda'}^0 | \mathcal{H}_{\delta}^{(1)} | u_{\lambda}^0 \rangle \right)
\end{aligned} \tag{165}$$

where the operators $\mathcal{H}_{\Delta}^{(1)}$ and $\mathcal{H}_{\delta}^{(1)}$ are defined such that $\langle u_{\lambda'}^0 | \mathcal{H}_{\Delta}^{(1)} | u_{\lambda}^0 \rangle$ is given by the first two terms, and $\langle u_{\lambda}^0 | \mathcal{H}_{\delta}^{(1)} | u_{\lambda}^0 \rangle$ by the last term, of Eq. (117) ($z \implies \zeta_l$ and ζ_l occur as subscripts in $\tilde{\Sigma}_{\zeta_l}^{(1)}$). With the aid of Eq. (161) and noting that the vector potential function used is in symmetric gauge, the second term of Eq. (164) can be shown to be equal to the third term. Putting all these results together, Eqs. (152), (160), and (163)-(165) in Eq. (150), we obtain the total susceptibility correction χ_{cor} as

χ_{cor}

$$\begin{aligned}
&= \sum_{\lambda} \left(\frac{1}{2\pi} \right)^3 \int d^3 k \, k_B T \sum_l 2 \left\langle u_{\lambda}^0(\vec{r}, \vec{k}, \zeta_l) \left| \int d^3 r' e^{i\vec{k} \cdot (\vec{r}' - \vec{r})} \tilde{\Sigma}_{\zeta_l}^{(2)}(\vec{r}, \vec{r}') u_{\lambda}^0(\vec{r}', \vec{k}, \zeta_l) \right\rangle \mathcal{G}_{\lambda}(\vec{k}, \zeta_l) \right. \\
&\quad + \sum_{\lambda} \left(\frac{1}{2\pi} \right)^3 \int d^3 k \, k_B T \sum_l \left\langle u_{\lambda}^0 | \mathcal{H}_{\delta}^{(1)} | u_{\lambda}^0 \right\rangle \tilde{H}_{\lambda}^{(1)}(\vec{k}, \zeta_l) \left[\mathcal{G}_{\lambda}(\vec{k}, \zeta_l) \right]^2 \\
&\quad + \sum_{\lambda} \left(\frac{1}{2\pi} \right)^3 \int d^3 k \, k_B T \sum_l 2 \left\langle u_{\lambda}^0(\vec{r}, \vec{k}, \zeta_l) \left| \int d^3 r' e^{i\vec{k} \cdot (\vec{r}' - \vec{r})} \tilde{\Sigma}_{\zeta_l}^{(1)}(\vec{r}, \vec{r}') \frac{e}{B\hbar c} A(\vec{r} - \vec{r}') \cdot \nabla_{\vec{k}} u_{\lambda}^0(\vec{r}', \vec{k}, \zeta_l) \right\rangle \right. \\
&\quad \quad \times \mathcal{G}_{\lambda}(\vec{k}, \zeta_l) \\
&\quad - \sum_{\lambda} \left(\frac{1}{2\pi} \right)^3 \int d^3 k \, k_B T \sum_l \sum_{\lambda' \neq \lambda} 2(\tilde{H}_{\lambda'}^0 - \tilde{H}_{\lambda}^0)^{-1} \langle u_{\lambda'}^0 | \mathcal{H}_{\delta}^{(1)} | u_{\lambda}^0 \rangle \langle u_{\lambda}^0 | \mathcal{H}_{\delta}^{(1)} | u_{\lambda'}^0 \rangle \mathcal{G}_{\lambda}(\vec{k}, \zeta_l) \\
&\quad - \sum_{\lambda} \left(\frac{1}{2\pi} \right)^3 \int d^3 k \, k_B T \sum_l \sum_{\lambda' \neq \lambda} (\tilde{H}_{\lambda'}^0 - \tilde{H}_{\lambda}^0)^{-1} \left(\langle u_{\lambda'}^0 | \mathcal{H}_{\Delta}^{(1)} | u_{\lambda}^0 \rangle \langle u_{\lambda}^0 | \mathcal{H}_{\delta}^{(1)} | u_{\lambda'}^0 \rangle \right. \\
&\quad \quad \left. + \langle u_{\lambda}^0 | \mathcal{H}_{\Delta}^{(1)} | u_{\lambda'}^0 \rangle \langle u_{\lambda'}^0 | \mathcal{H}_{\delta}^{(1)} | u_{\lambda}^0 \rangle \right) \mathcal{G}_{\lambda}(\vec{k}, \zeta_l)
\end{aligned} \tag{166}$$

The second term gives a correction to χ_{CP} and the rest are corrections to χ_{ID} . We shall see that these corrections to χ_{CP} and χ_{ID} lead, among other things, to the cancellation of the appearance of quadratic terms in $\tilde{\Sigma}_{\zeta_l}^{(1)}$ as well as the total cancellation of the appearance of $\tilde{\Sigma}_{\zeta_l}^{(2)}$. This important cancellation is expected and is in agreement with the work of Philippas and McClure.⁶⁵ Using Eqs. (117) and (118) to write down χ_{CP} and χ_{ID} explicitly and denoting the corrected χ_{CP} and χ_{ID} by χ_{CP}^{Σ} and χ_{ID}^{Σ} , respectively, we may write the

total magnetic susceptibility of interacting free and Bloch electrons as

$$\chi = \chi_{LP} + \chi_{CP}^{\Sigma} + \chi_{ID}^{\Sigma} \quad (167)$$

χ_{LP} is given by Eq. (145), χ_{CP}^{Σ} and χ_{ID}^{Σ} are given by the following relations:

$$\chi_{CP}^{\Sigma} = - \sum_{\lambda} \left(\frac{1}{2\pi} \right)^3 \int d^3k \, k_B T \sum_l \left\{ \left[\tilde{H}_{\Delta,\lambda}^{(1)}(\vec{k}, \zeta_l) \right]^2 + \tilde{H}_{\Delta,\lambda}^{(1)}(\vec{k}, \zeta_l) \tilde{H}_{\delta,\lambda}^{(1)}(\vec{k}, \zeta_l) \right\} \left[\mathcal{G}_{\lambda}(\vec{k}, \zeta_l) \right]^2 \quad (168)$$

$$\chi_{ID}^{\Sigma} = - \sum_{\lambda} \left(\frac{1}{2\pi} \right)^3 \int d^3k \, k_B T \sum_l 2W_{\lambda}^{(2)}(\vec{k}, \zeta_l) \mathcal{G}_{\lambda}(\vec{k}, \zeta_l) \quad (169)$$

where

$$\begin{aligned} \tilde{H}_{\Delta,\lambda}^{(1)}(\vec{k}, \zeta_l) &= \left\langle u_{\lambda}^0 \left| \mathcal{H}_0^{(1)} \frac{e}{B\hbar c} \left[A(\nabla_{\vec{k}}) \tilde{H}_{\lambda}^0(\vec{k}, \zeta_l) \right] \cdot \nabla_{\vec{k}} u_{\lambda}^0 \right\rangle \\ &\quad + \left\langle u_{\lambda}^0(\vec{r}, \vec{k}, \zeta_l) \left| \int d^3r' e^{i\vec{k} \cdot (\vec{r}' - \vec{r})} \tilde{\Sigma}_{\zeta_l}^0(\vec{r}, \vec{r}') \frac{e}{B\hbar c} A(\vec{r} - \vec{r}') \cdot \nabla_{\vec{k}} u_{\lambda}^0(\vec{r}', \vec{k}, \zeta_l) \right\rangle \right. \end{aligned} \quad (170)$$

$$\tilde{H}_{\delta,\lambda}^{(1)}(\vec{k}, \zeta_l) = \left\langle u_{\lambda}^0(\vec{r}, \vec{k}, \zeta_l) \left| \int d^3r' e^{i\vec{k} \cdot (\vec{r}' - \vec{r})} \tilde{\Sigma}_{\zeta_l}^{(1)}(\vec{r}, \vec{r}') u_{\lambda}^0(\vec{r}', \vec{k}, \zeta_l) \right\rangle = \Sigma_{\lambda\lambda}^{(1)0}(\vec{k}, \zeta_l) \quad (171)$$

$$\begin{aligned} W_{\lambda}^{(2)}(\vec{k}, \zeta_l) &= \left\langle u_{\lambda}^0 \left| \mathcal{H}_0^{(2)} \right| u_{\lambda}^0 \right\rangle + \left\langle u_{\lambda}^0 \left| \mathcal{H}_0^{(1)} \right| u_{\lambda}^{(1)} \right\rangle - \left\langle u_{\lambda}^0 \left| \sum_{\vec{q}} e^{i\vec{k} \cdot \vec{q}} \tilde{H}_{\lambda}^0(\vec{q}, \zeta_l) \frac{1}{2!} \left(\frac{e}{B\hbar c} A(\vec{q}) \cdot \nabla_{\vec{k}} \right)^2 \right| u_{\lambda}^0 \right\rangle \\ &\quad + \left\langle u_{\lambda}^0 \left| \frac{e}{B\hbar c} A(\nabla_{\vec{k}}) \tilde{H}_{\lambda}^0(\vec{k}, \zeta_l) \cdot i\nabla_{\vec{k}} \right| u_{\lambda}^{(1)} \right\rangle + \left\langle u_{\lambda}^0 \left| \frac{e}{B\hbar c} A(\nabla_{\vec{k}}) \tilde{H}_{\lambda}^{(1)}(\vec{k}, \zeta_l) \cdot i\nabla_{\vec{k}} \right| u_{\lambda}^0 \right\rangle \\ &\quad - \tilde{H}_{\Delta,\lambda}^{(1)}(\vec{k}, \zeta_l) \frac{1}{4} \frac{e}{\hbar c} \left(\frac{\partial}{\partial k_y} \left\langle u_{\lambda}^0 \left| i \frac{\partial}{\partial k_x} u_{\lambda}^0 \right\rangle - \frac{\partial}{\partial k_x} \left\langle u_{\lambda}^0 \left| i \frac{\partial}{\partial k_y} u_{\lambda}^0 \right\rangle \right) \right) \\ &\quad + \left\langle u_{\lambda}^0(\vec{r}, \vec{k}, \zeta_l) \left| \int d^3r' e^{i\vec{k} \cdot (\vec{r}' - \vec{r})} \tilde{\Sigma}_{\zeta_l}^0(\vec{r}, \vec{r}') \frac{1}{2!} \left(\frac{e}{B\hbar c} A(\vec{r} - \vec{r}') \cdot \nabla_{\vec{k}} \right)^2 u_{\lambda}^0(\vec{r}', \vec{k}, \zeta_l) \right\rangle \right. \\ &\quad + \left\langle u_{\lambda}^0(\vec{r}, \vec{k}, \zeta_l) \left| \int d^3r' e^{i\vec{k} \cdot (\vec{r}' - \vec{r})} \tilde{\Sigma}_{\zeta_l}^0(\vec{r}, \vec{r}') \left(\frac{e}{B\hbar c} A(\vec{r} - \vec{r}') \cdot \nabla_{\vec{k}} \right) u_{\lambda}^{(1)}(\vec{r}', \vec{k}, \zeta_l) \right\rangle \right. \\ &\quad \left. - \frac{1}{2} \sum_{\lambda' \neq \lambda} \left(\tilde{H}_{\lambda'}^0 - \tilde{H}_{\lambda}^0 \right)^{-1} \left(\left\langle u_{\lambda'}^0 \left| \mathcal{H}_{\Delta}^{(1)} \right| u_{\lambda}^0 \right\rangle \left\langle u_{\lambda}^0 \left| \mathcal{H}_{\delta}^{(1)} \right| u_{\lambda'}^0 \right\rangle - \left\langle u_{\lambda}^0 \left| \mathcal{H}_{\Delta}^{(1)} \right| u_{\lambda'}^0 \right\rangle \left\langle u_{\lambda'}^0 \left| \mathcal{H}_{\delta}^{(1)} \right| u_{\lambda}^0 \right\rangle \right) \end{aligned} \quad (172)$$

Indeed, χ is a linear function of the operator $\tilde{\Sigma}_{\zeta_l}^{(1)}$ and is independent of $\tilde{\Sigma}_{\zeta_l}^{(2)}$. For reasons which maybe clarified in some well-known cases, we will refer to the $\tilde{\Sigma}_{\zeta_l}^{(1)}$ term in χ_{CP}^{Σ} as the “enhancement term”. Consequently, we will also refer to the $\tilde{\Sigma}_{\zeta_l}^{(1)}$ terms in χ_{ID}^{Σ} as the “second-order effect of the enhancement”.

I. Application of χ to Some Many-Body Systems

The general formula will be applied to (a) a Fermi liquid and to (b) correlated electrons represented by the Hubbard model,⁶⁶ in Hartree-Fock approximation for simplicity. In what follows, electric charge $e \implies -e$.

1. Fermi liquid

Since the periodic wave function $u_\lambda^0(r, k, \zeta_l)$ occurring in Eqs. (170) and (172) is a constant quantity for Fermi liquids, we can immediately write down the magnetic susceptibility of the quasiparticles as

$$\chi = \chi_{LP} + \chi_{CP}^\Sigma \quad (173)$$

We obtain using Eqs. (4.62), (4.67), and (4.69) of Ref.⁶⁶, the total quasiparticle energy [more appropriately the Weyl transform with $\vec{p} + (e/c)\vec{A}(\vec{q}) \Rightarrow \hbar\vec{k}$] in a magnetic field as

$$\tilde{H}_\lambda(\vec{k}, B, \zeta_l) = e^0(\vec{k}) \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix} + B\mu_B \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix} + \frac{B\chi_{CP}^\Sigma}{\mu_B} \frac{\pi^2 \hbar^2}{m^* k_F} B_0 \begin{pmatrix} -1 & 0 \\ 0 & 1 \end{pmatrix} \quad (174)$$

where $m^* = (1 + \frac{1}{3}A_1)m$; A_1 and B_0 are well-known Fermi-liquid parameters. We immediately identify, upon examination of Eqs. (130), (170), and (171), the following relations:

$$\tilde{H}_\lambda^0(k, \zeta_l) = e^0(k) \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix} \quad (175)$$

$$\tilde{H}_{\Delta, \lambda}^{(1)}(k, \zeta_l) = \mu_B \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix} \quad (176)$$

$$\tilde{H}_{\delta, \lambda}^{(1)}(k, \zeta_l) = \frac{\chi_{CP}^\Sigma}{\mu_B} \frac{\pi^2 \hbar^2}{m^* k_F} B_0 \begin{pmatrix} -1 & 0 \\ 0 & 1 \end{pmatrix} \quad (177)$$

Substituting these quantities in χ_{CP}^Σ , Eq. (168), we get

$$\chi_{CP}^\Sigma = \left[\left(\frac{1 + \frac{1}{3}A_1}{1 + B_0} \right) \right] \chi_P^0 \quad (178)$$

where χ_P^0 is the Pauli spin susceptibility for a noninteracting electron gas. The calculation of χ_{LP} is very elementary and the total χ is thus given by

$$\chi = \left(1 + \frac{1}{3}A_1 \right)^{-1} \chi_{LP}^0 + \left[\left(\frac{1 + \frac{1}{3}A_1}{1 + B_0} \right) \right] \chi_P^0 \quad (179)$$

This is a very well-known result for the orbital and spin susceptibility of Fermi liquids. Note that a small effective mass enhances χ_{LP}^0 .

2. Hubbard model in Hartree-Fock approximation

The model under consideration assumes that there is only one band of interest energetically far removed from the other bands. For a very narrow band we may write

$$\chi \simeq \chi_{CP}^{\Sigma} + \chi_{ID}^{\Sigma} \quad (180)$$

Upon transforming Eqs. (4.75) and (4.76) of Ref.⁶⁶ to k space, we have for the expression of the total Hubbard Hamiltonian in a magnetic field in the Hartree-Fock approximation as

$$H = \sum_{\vec{k}, \sigma} e(\vec{k}) n(\vec{k}, \sigma) + \sum_{\vec{k}, \sigma} I \langle n(\vec{k}, \sigma) \rangle n(\vec{k}, -\sigma) + \frac{1}{2} g \mu_B B \sum_{\vec{k}} [n(\vec{k}, \uparrow) - n(\vec{k}, \downarrow)] \quad (181)$$

Therefore, $\tilde{H}_{\lambda}(\vec{k}, \zeta_l, B)$ is given by

$$\tilde{H}_{\lambda}(\vec{k}, \zeta_l, B) = e(\vec{k}) \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix} + I \begin{pmatrix} \langle n(\vec{k}, \downarrow) \rangle & 0 \\ 0 & \langle n(\vec{k}, \uparrow) \rangle \end{pmatrix} + \frac{1}{2} g \mu_B B \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix} \quad (182)$$

In view of the fact that $\langle n(\vec{k}, \downarrow) \rangle$ is greater than $\langle n(\vec{k}, \uparrow) \rangle$, we may write

$$\langle n(\vec{k}, \downarrow) \rangle = n + \delta n \quad (183)$$

$$\langle n(\vec{k}, \uparrow) \rangle = n - \delta n \quad (184)$$

$$\frac{2N\delta n}{V} \frac{1}{2} g \mu_B = \chi_{CP} B \quad (185)$$

and readily obtain

$$\tilde{H}_{\lambda}^0(k, \zeta_l) = [e^0(k) + In] \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix} \quad (186)$$

$$\tilde{H}_{\Delta, \lambda}^{(1)}(k, \zeta_l) = \frac{1}{2} g \mu_B \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix} \quad (187)$$

$$\tilde{H}_{\delta, \lambda}^{(1)}(k, \zeta_l) = I \left(\frac{V}{N} \right) \frac{\chi_{CP}}{g \mu_B} \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix} \quad (188)$$

Upon substitution of these quantities in Eq. (168), we obtain

$$\chi_{CP}^{\Sigma} = \chi_0 \left(1 - 2I \left(\frac{V}{N} \right) \frac{\chi_0}{(g \mu_B)^2} \right)^{-1} \quad (189)$$

leading to the Stoner criterion for the appearance of ferromagnetism.⁶⁶

To obtain χ_{ID}^{Σ} , we note that in Eq. (172)

$$W_{\lambda}^{(2)}(\vec{k}, \zeta_l) \simeq \langle u_{\lambda}^0(\vec{r}, \vec{k}) | \mathcal{H}_0^{(2)} | u_{\lambda}^0(\vec{r}, \vec{k}) \rangle. \quad (190)$$

The second term, representing a Van Vleck paramagnetism, and last term of Eq. (172) are neglected since the band of interest is energetically far removed from other bands. The third up to sixth term, inclusive, are neglected by the assumption of a very narrow band and the rest of Eq. (172) is neglected due to the δ -function locality of $\tilde{\Sigma}_{\zeta_l}^0(\vec{r}, \vec{r}')$. Expressing $e^{ik \cdot r} u_{\lambda}(r, k, \zeta_l)$ as a linear combination of atomic orbitals we obtain, upon substitution in Eq. (169), a familiar “atomic diamagnetism” multiplied by the total number of electrons N in the band

$$\chi_{ID}^{\Sigma} = - \left(\frac{Ne^2}{4mc^2} \right) \langle \phi_{\lambda}(r) | x^2 + y^2 | \phi_{\lambda}(r) \rangle \quad (191)$$

where $\phi_{\lambda}(r)$ is the atomic orbital of the band. For most purposes χ_{ID}^{Σ} is neglected and $\chi \simeq \chi_{CP}^{\Sigma}$.

VII. MAGNETIC SUSCEPTIBILITY OF DILUTE NONMAGNETIC ALLOYS

Theoretical efforts toward giving a general expression of the magnetic susceptibility χ for solids with nonmagnetic impurities were initiated by Kohn and Luttinger⁶⁷ by considering an idealized model of free-electron band. Other attempts to give χ for general Bloch bands can at best proceed only as a power-series expansion in the strength of the impurity potential. However, experimental data indicate the need for a better understanding and a more complete theory that incorporates the band-structure effects of the host lattice.

A. Lattice Weyl-Wigner Formalism Approach

We approach the problem by the use of the lattice Weyl-Wigner formalism of quantum theory, not very widely known in solid-state physics, although its embryonic and disguised form is already apparent in the operator method of Roth⁵⁴ and Blount⁸ and in the formalism of the dynamics of band electrons by Wannier¹. The result for χ is given to order \hbar^2 valid for general nondegenerate Bloch bands and to all orders in the impurity potential. The effect of Bloch-electron interaction can, in principle, be incorporated by the use of a screened impurity potential. The expression for χ reduces to all well-known limiting cases. It is applied to the free-electron-band model of dilute alloys of copper. The result gives a firm theoretical foundation to the empirical theory of Henry and Rogers, which accounts quite well of their experimental results.

The final result for χ , the change of the magnetic susceptibility of the crystalline solid due to

the presence of impurity centers, may be written as,

$$\Delta\chi = N_I(\chi - \chi_0)$$

where N_I is the number of impurity centers, χ_0 is the magnetic susceptibility of the pure crystal host, and χ is given by the following formula:

$$\chi = -\frac{1}{V} \left(\frac{1}{h} \right)^3 \text{Tr} \int d^3p d^3q \left(-\frac{\hbar^2}{48} \left\{ \begin{aligned} & \frac{\partial f(\Sigma^0)}{\partial \Sigma^0} (\Sigma^{(1)})^2 + f(\Sigma^0) \Sigma^{(2)} \\ & \frac{\partial f(\Sigma^0)}{\partial \Sigma^0} \left(\begin{aligned} & \left[\frac{\partial^2 \Sigma^{(2)}}{\partial \vec{p} \partial \vec{p}}; \frac{\partial^2 V^0}{\partial \vec{q} \partial \vec{q}} \right] + 2 \left[\frac{\partial^2 \Sigma^{(1)}}{\partial \vec{p} \partial \vec{p}}; \frac{\partial^2 V^{(1)}}{\partial \vec{q} \partial \vec{q}} \right] \\ & + \left[\frac{\partial^2 \Sigma^0}{\partial \vec{p} \partial \vec{p}}; \frac{\partial^2 \Sigma^{(2)}}{\partial \vec{q} \partial \vec{q}} \right] - \left[\frac{\partial^2 \Sigma^{(2)}}{\partial \vec{p} \partial \vec{q}}; \frac{\partial^2 V^0}{\partial \vec{q} \partial \vec{p}} \right] \\ & - 2 \left[\frac{\partial^2 \Sigma^{(1)}}{\partial \vec{p} \partial \vec{q}}; \frac{\partial^2 \Sigma^{(1)}}{\partial \vec{q} \partial \vec{p}} \right] - \left[\frac{\partial^2 V^0}{\partial \vec{p} \partial \vec{q}}; \frac{\partial^2 \Sigma^{(2)}}{\partial \vec{q} \partial \vec{p}} \right] \end{aligned} \right) \\ & \Sigma^{(2)} \frac{\partial^2 f(\Sigma^0)}{(\partial \Sigma^0)^2} \left(\left[\frac{\partial^2 \Sigma^0}{\partial \vec{p} \partial \vec{p}}; \frac{\partial^2 V^0}{\partial \vec{q} \partial \vec{q}} \right] - \left[\frac{\partial^2 V^0}{\partial \vec{p} \partial \vec{q}}; \frac{\partial^2 V^0}{\partial \vec{q} \partial \vec{p}} \right] \right) \\ & + (\Sigma^{(1)})^2 \frac{\partial^3 f(\Sigma^0)}{(\partial \Sigma^0)^3} \left(\left[\frac{\partial^2 \Sigma^0}{\partial \vec{p} \partial \vec{p}}; \frac{\partial^2 V^0}{\partial \vec{q} \partial \vec{q}} \right] - \left[\frac{\partial^2 V^0}{\partial \vec{p} \partial \vec{q}}; \frac{\partial^2 V^0}{\partial \vec{q} \partial \vec{p}} \right] \right) \\ & + 2 \Sigma^{(1)} \frac{\partial^2 f(\Sigma^0)}{(\partial \Sigma^0)^2} \left(\begin{aligned} & \left[\frac{\partial^2 \Sigma^{(1)}}{\partial \vec{p} \partial \vec{p}}; \frac{\partial^2 V^0}{\partial \vec{q} \partial \vec{q}} \right] + \left[\frac{\partial^2 \Sigma^0}{\partial \vec{p} \partial \vec{p}}; \frac{\partial^2 V^{(1)}}{\partial \vec{q} \partial \vec{q}} \right] \\ & - \left[\frac{\partial^2 \Sigma^{(1)}}{\partial \vec{p} \partial \vec{q}}; \frac{\partial^2 V^0}{\partial \vec{q} \partial \vec{p}} \right] - \left[\frac{\partial^2 V^0}{\partial \vec{p} \partial \vec{q}}; \frac{\partial^2 \Sigma^{(1)}}{\partial \vec{q} \partial \vec{p}} \right] \end{aligned} \right) \end{aligned} \right\} \right) \quad (192)$$

For magnetic field in the z -direction using a symmetric gauge, the various quantities entering in the above expression are defined as follows: $f(x)$ is the Fermi-dirac distribution function, and

$$\Sigma^0 = W^0(\vec{p}) + V^0(\vec{p}, \vec{q}), \quad (193)$$

$$\Sigma^{(1)} = \left(\frac{e}{2c} \right) (\vec{q} \times \nabla_{\vec{p}})_z (\vec{q} \times \nabla_{\vec{p}})_z W^0(\vec{p}) + W^{(1)}(\vec{p}) + V^{(1)}(\vec{p}, \vec{q}), \quad (194)$$

$$\begin{aligned} \Sigma^{(2)} = & \left(\frac{e}{2c} \right)^2 (\vec{q} \times \nabla_{\vec{p}})_z (\vec{q} \times \nabla_{\vec{p}})_z W^0(\vec{p}) + \left(\frac{e}{c} \right) (\vec{q} \times \nabla_{\vec{p}})_z W^{(1)}(\vec{p}) \\ & + 2W^{(2)}(\vec{p}) + 2V^{(2)}(\vec{p}, \vec{q}) \end{aligned} \quad (195)$$

For simplicity one may take $V^0(\vec{p}, \vec{q})_{\lambda\lambda'} = V_{\lambda}^0(\vec{p}, \vec{q}) \delta_{\lambda\lambda'}$, making Σ^0 diagonal in bands. Equation (192) has the novel features of being transparent and of being valid to all orders in the impurity potential for general nondegenerate Bloch bands.

B. Application to Dilute Alloys of Copper

We will show that in the free-electron-band model Eq. (192) gives a firm theoretical foundation of the empirical theory of Henry and Rogers⁶⁸ which accounts quite well for their experimental results for various solutes in copper. This is in marked contrast to the theory

of Kohn and Luming⁶⁷ which fails to justify the formula for $\Delta\chi$ per solute atom used by Henry and Rogers.

For the free-electrons in cooper, we have

$$\Sigma^0 = \frac{p^2}{2m} + V_I(\vec{q}), \quad (196)$$

$$\Sigma^{(1)} = \left(\frac{e}{2mc}\right)(\vec{q} \times \vec{p})_z + \mu_B \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}, \quad (197)$$

$$\Sigma^{(2)} = \left(\frac{e}{2c}\right)^2 \frac{1}{m} (q_x^2 + q_y^2). \quad (198)$$

Thus, Eq. (192) reduces to

$$\chi = \chi_1 + \chi_2 + \chi_3 + \chi_4 + \chi_5 \quad (199)$$

where

$$\chi_1 = -2 \frac{1}{Vh^3} \int d^3p d^3q f'(\Sigma^0) \left[\left(\frac{e}{2mc} L_z \right)^2 + \mu_B^2 \right], \quad (200)$$

$$\chi_2 = -2 \frac{1}{Vh^3} \int d^3p d^3q f(\Sigma^0) \frac{e^2}{4mc^2} (q_x^2 + q_y^2), \quad (201)$$

$$\chi_3 = 2 \frac{1}{Vh^3} \int d^3p d^3q f'(\Sigma^0) \mu_B^2, \quad (202)$$

$$\chi_4 = 2 \frac{1}{24Vh^3} \int d^3p d^3q f'''(\Sigma^0) \frac{\hbar^2}{m} \nabla^2 V_I(q) \left[\left(\frac{e}{2mc} L_z \right)^2 + \mu_B^2 \right], \quad (203)$$

$$\chi_5 = 2 \frac{1}{24Vh^3} \int d^3p d^3q f''(\Sigma^0) \frac{\hbar^2}{m} \nabla^2 V_I(q) \frac{e^2}{4mc^2} (q_x^2 + q_y^2) \quad (204)$$

where factors of 2 in front of integrals account for the \pm spin band. We note that for $V_I(q) = 0$, we have

$$\chi = \chi^0 = \chi_{spin}^0 + \chi_{orb}^0$$

where

$$\chi_{orb}^0 = \frac{2}{3Vh^3} \int d^3p d^3q f' \left(\frac{p^2}{2m} \right) \mu_B^2, \quad (205)$$

$$\chi_{spin}^0 = -\frac{2}{Vh^3} \int d^3p d^3q f' \left(\frac{p^2}{2m} \right) \mu_B^2. \quad (206)$$

In view of Eqs. (200), (202), and (203), and Eqs. (205) and (206), we can write for an arbitrary strength of $V_I(q)$,

$$\Delta\chi_{orb} = \chi_{orb}^0 \frac{\Delta g_1}{g}, \quad (207)$$

$$\Delta\chi_{spin} = \chi_{spin}^0 \frac{\Delta g_2}{g}, \quad (208)$$

where

$$g = \frac{2}{Vh^3} \int d^3p d^3q f' \left(\frac{p^2}{2m} \right), \quad (209)$$

$$\Delta g_1 = \frac{2}{Vh^3} \int d^3p d^3q \left[f'(\Sigma^0) - f' \left(\frac{p^2}{2m} \right) \right], \quad (210)$$

$$\Delta g_2 = \Delta g_1 + \frac{2}{24Vh^3} \int d^3p d^3q f'''(\Sigma^0) \frac{\hbar^2}{m} \nabla^2 V_I(q) \quad (211)$$

By writing $(L_z)^2 = (q \times p)_z^2 = q_x^2 p_y^2 - 2q_x q_y p_x p_y + q_y^2 p_x^2$ and integrating with respect to \vec{p} , the first terms of χ_1 , and χ_4 can be combined with χ_2 and χ_5 , resulting in the expression for $\Delta\chi$ per solute atom as

$$\Delta\chi = -\frac{e^2}{6mc^2} \int d^3q \Delta\rho(\vec{q}) |q|^2 + \chi_{orb}^0 \frac{\Delta g_1}{g} + \chi_{spin}^0 \frac{\Delta g_2}{g}, \quad (212)$$

where

$$\begin{aligned} \Delta\rho(\vec{q}) = & \frac{2}{Vh^3} \int d^3p \left\{ \left[f(\Sigma^0) + \frac{1}{24} f''(\Sigma^0) \frac{\hbar^2}{m} \nabla^2 V_I(q) - f \left(\frac{p^2}{2m} \right) \right] + \left[f'(\Sigma^0) \right. \right. \\ & \left. \left. + \frac{1}{24} f'''(\Sigma^0) \frac{\hbar^2}{m} \nabla^2 V_I(q) - f' \left(\frac{p^2}{2m} \right) \right] \frac{p^2}{3m} \right\} \end{aligned} \quad (213)$$

Equation (212), with $\Delta g_1 = \Delta g_2$ and a similar consistent approximation for $\rho(\vec{q})$, is exactly the expression used by Henry and Rogers,⁶⁸ as pointed out by Kohn and Luming⁶⁷, in analyzing their data on dilute alloys of Zn, Ga, Ge, and As with Cu which accounts quite well for their experimental results. Thus the use of Eq. (212) by Henry and Rogers, as pointed out by Kohn and Luming, is given a firm theoretical foundation. Here lies the essential discrepancy between Eq. (212) and the theory of Kohn and Luming. We believe that the copper conduction electron can be approximately described by a free electron-band model and Eq. (192) should provide a good approximation for copper as used by Henry and Rogers. On the other hand, the theory presented by Kohn and Luming does not contain the entire emperical expression for $\Delta\chi$ per solute atom used by Henry and Rogers.⁶⁸

VIII. MAGNETIC SUSCEPTIBILITY OF DILUTE MAGNETIC ALLOYS

For completeness, we will briefly treat the magnetic susceptibility of dilute magnetic alloys. The study of dilute magnetic alloys evolves with the general problem of how magnetism develops in magnets. As it turns out the study of dilute magnetic alloys has stood

out as a complex many-body problem dealing with self-consistent fluctuating scattering potential, and hence becomes an important physics problem in its own right. Self-consistency in the sense that the electrons interacting with the impurity themselves create the fluctuating potential, signaling a *bonafide* many-body and perhaps a time-dependent and/or highly nonlinear problem. Roughly speaking, each electron as it passes the impurity influences the state of the impurity and is influenced by the impurity. Therefore, the state of the ion which a given electron sees is determined by all previous electron-impurity encounters. The problem is essentially a many-body problem with nonstationary impurity potential.

The increase in resistance due to strong fluctuations at low temperature is reminiscent of the Anderson localization problem with random scattering potential. Here, the magnetic impurities are represented by localized spins that couple to the conduction-band electrons of the nonmagnetic host metal via a spin-exchange interaction, in particular via anti-ferromagnetic coupling. Whether the magnetic moment of the impurity persists down to zero temperature is not very well understood and this has given way to the so-called Kondo problem, where one studies the low-temperature behavior of a system.

At sufficiently low impurity density we may concentrate on a single impurity localized, say, at $x = 0$ and study how its magnetic properties are modified due to its coupling with the electrons. A rough hand-waving argument of the physics of the system may be made based on the time scale of observation versus the spin-flip scattering or spin relaxation time. Denoting the spin-flip relaxation time by τ and the time scale of observation by $\Delta t = \frac{\hbar}{k_B T}$, where k_B is the Boltzmann constant and T is the temperature, then when $\Delta t \ll \tau$, the spin orientation almost remains constant during the time τ . This condition defines the weak coupling regime (note that Schrieffer⁶⁹ use the interaction time, τ_U , instead of Δt of Kondo⁷⁰). On the other hand when $\Delta t \gg \tau$, then spin-flip becomes frequent and the up and down orientation of spin appear equally, i.e., there is a strong fluctuation of impurity potential due to frequent spin flip during the time Δt . This condition defines the strong coupling regime, it is referred to as the screening or *quenching* of the impurity spin since one observes vanishing impurity spin, leading to finite susceptibility at $T = 0$. There is a temperature region where $\Delta t \simeq \tau$, the corresponding temperature is often referred to as the Kondo temperature, denoted by T_K , after Jun Kondo who first study the dilute magnetic alloy problem in 1964. For $T < T_K$, we have the strong coupling regime while for $T > T_K$, we have the weak coupling regime, as shown in Fig. 3. The magnetic susceptibility changes

from Curie behavior for $T > T_K$ to Pauli spin susceptibility for $T < T_K$. Thus, the impurity is said to have a moment if the susceptibility due to the impurity shows a $\frac{1}{k_B T}$ dependence (such as χ_{spin} in Eq. (206) down to $T = 0$).

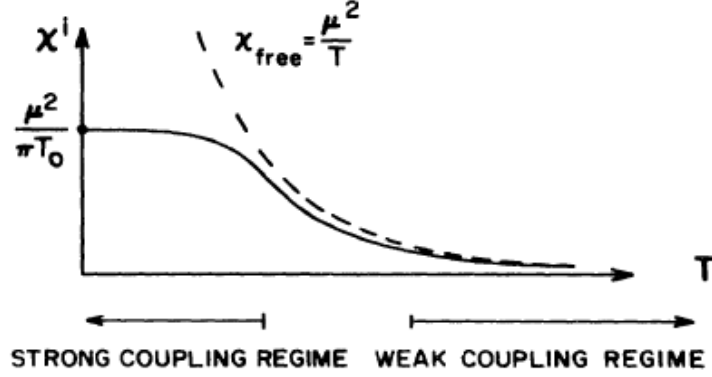


FIG. 3: The *impurity* susceptibility χ^i is shown compared with the free-spin susceptibility χ_{free} . At high temperatures χ^i approaches χ_{free} logarithmically on the scale set by T_K . As the temperature is lowered, it goes to a finite value at $T = 0$, indicative of a screened spin. Reproduced after Ref.⁷¹

A. States of Magnetic Impurity in Nonmagnetic Metal Host

Here we consider only the Hamiltonian of impurity in nonmagnetic metals and see how the *localized* spin can be generated by the interaction with conduction electrons. Following Kondo,⁷⁰ we write the ‘bare’ Hamiltonian as

$$H = \sum_{\vec{k}\sigma} \varepsilon_{\vec{k}} a_{\vec{k}\sigma}^\dagger a_{\vec{k}\sigma} + V_0 \sum_{\vec{k}\sigma} (a_{\vec{k}\sigma}^\dagger a_{0\sigma} + a_{0\sigma}^\dagger a_{\vec{k}\sigma}) + \varepsilon_0 \sum_{\sigma} a_{0\sigma}^\dagger a_{0\sigma} + U a_{0\uparrow}^\dagger a_{0\uparrow} a_{0\downarrow}^\dagger a_{0\downarrow} \quad (214)$$

where $\varepsilon_{\vec{k}}$ is the conduction energy-band function, ε_0 is the impurity orbital $l = 0$ energy level, U is the Coulomb interaction energy between electrons, and V_0 is the self-consistent potential. Note that V_0 is sufficient enough for fixing the values of the impurity levels if U is not large, i.e., the problem then is simply a one-body problem. However, if U is large the problem becomes a complex many-body problem.

B. Generation of localized moment

Consider the U term in Eq. (214). If the spin \uparrow electron fills the localized orbital, i.e., $a_{0\uparrow}^\dagger a_{0\uparrow} = 1$ then the last two terms reduces to

$$\varepsilon_0 \sum_{\sigma} a_{0\sigma}^\dagger a_{0\sigma} + U a_{0\uparrow}^\dagger a_{0\uparrow} a_{0\downarrow}^\dagger a_{0\downarrow} \implies (\varepsilon_0 + U) a_{0\downarrow}^\dagger a_{0\downarrow}$$

This shows that the electron down spin state is raised up in energy by the Coulomb interaction U . If it happens that $(\varepsilon_0 + U) > \varepsilon_F$ then $\langle a_{0\downarrow}^\dagger a_{0\downarrow} \rangle \implies 0$ and *localized* \uparrow -spin is generated. The same argument holds if \uparrow and \downarrow are interchanged. On the other hand localized spin does not emerge for the following two cases: (a) $\{\varepsilon_0, (\varepsilon_0 + U)\} < \varepsilon_F$ and (b) $\{\varepsilon_0, (\varepsilon_0 + U)\} > \varepsilon_F$.

1. Fluctuating localized moment

The localized spin if present is actually fluctuating by virtue of the spin-exchange process. A spin \downarrow electron from the conduction band may fall into the localized orbital, which has been filled by the spin \uparrow electron from the conduction band. Then an exchange process occurs in which the spin \uparrow electron goes back to the conduction band thereby resulting in an overall spin exchange and hence a localized \downarrow spin emerges, and vice versa. This is best illustrated for quantum dots as shown in Fig. 4. Note in magnetic alloys, a complementary process corresponding to that depicted for quantum dots in Fig. 4, can also occur in which the spin \downarrow electron from the conduction band leap to the level $\varepsilon_0 + U$ of the localized orbital, then fall down the level ε_0 kicking the spin \downarrow electron back to the conduction band, again resulting in overall spin exchange.⁷⁰ The most likely event between the two processes of course depends on the magnitude of the Coulomb energy U as well as on ε_0 , as can be seen by simply sliding down the U and ε_0 (keeping their distance fixed) in Fig. 4.

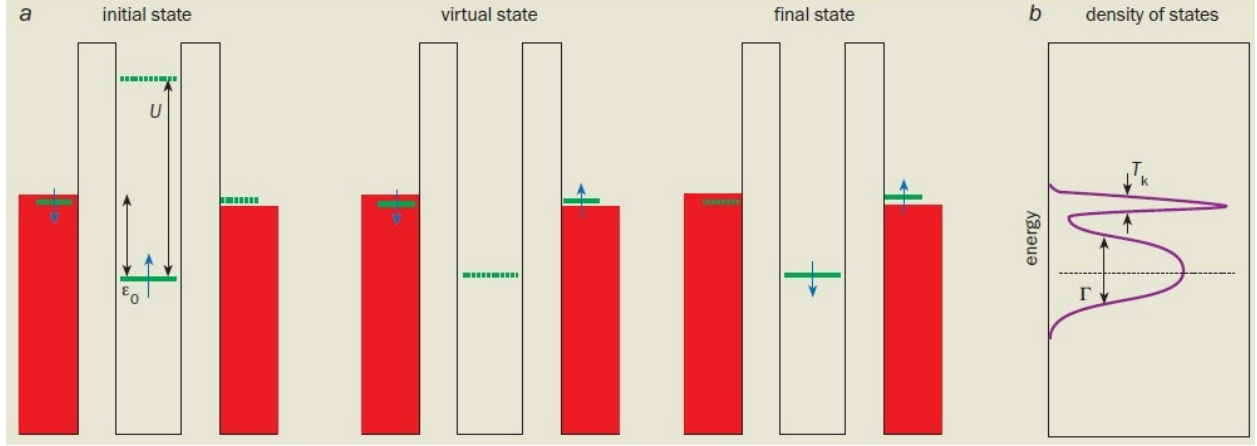


FIG. 4: Spin flip process: (a) Anderson model assumes just one electron level, ε_0 , below the Fermi energy of the metal. This level is occupied by spin \uparrow electron. Adding another electron is prohibited by the Coulomb energy, U . It also cost energy, ε_0 , to remove the electron from the impurity (assuming Fermi level is set to zero). However, quantum mechanically, the spin up electron may tunnel out of the impurity site to briefly occupy a classically forbidden ‘virtual state’ outside the impurity. Then this is replaced by the electron from the metal. This effectively flip the spin of the impurity. (b) Many such events combine to produce the appearance of of an extra resonance at the Fermi energy. This extra resonance can remarkably change the conductance. [Figure reproduce from Ref.⁷²]

Whether indeed the localized spin is effectively present or not depends on the time scale of interaction, also referred to by Kondo⁷⁰ as the time scale of observation. This time scale is determined by the width of the localized energy level, designated as Δ_0 . If the fluctuation occurs much more slowly than the observation time, then the localized spin is resolved or is present. On the contrary, if the observation time is sufficiently greater than the spin fluctuation, say at low temperature, then the localized spin is no longer resolve and appears to have vanished. This often referred to as screened or compensated localized moment.

C. The s - d interaction

In the Kondo problem, it is the two-electron interaction U that makes the fluctuating localized moment and brings the problem to a higher degree of difficulty. When the two-electron interaction U is large and is dominant in the problem, Kondo was able to obtain an

effective Hamiltonian with the so-called s - d interaction terms, H_{sd} , where H_{sd} is given by

$$H_{sd} = -J \sum_{\vec{k}\vec{k}'} \left[S_z \left(a_{\vec{k}'\uparrow}^\dagger a_{\vec{k}\uparrow} - a_{\vec{k}'\downarrow}^\dagger a_{\vec{k}\downarrow} \right) + S_+ a_{\vec{k}'\downarrow}^\dagger a_{\vec{k}\uparrow} + S_- a_{\vec{k}'\uparrow}^\dagger a_{\vec{k}\downarrow} \right] + V \sum_{\vec{k}\vec{k}'\sigma} a_{\vec{k}'\sigma}^\dagger a_{\vec{k}\sigma}$$

where

$$J = V_0^2 \left(\frac{1}{\varepsilon_0} - \frac{1}{\varepsilon_0 + U} \right) < 0$$

$$V = -\frac{V_0^2}{2} \left(\frac{1}{\varepsilon_0} + \frac{1}{\varepsilon_0 + U} \right)$$

The term proportional to J is called the s - d interaction.

D. Bethe *Ansatz* Treatment of Exact Solution: Chiral Gross-Neveu Model

The Kondo problem has been interesting both in its own right where powerful mathematical techniques and other ideas have been tested for its solution. It has been approached by various theories, namely, perturbation theory, various resummation techniques, S-matrix formalism, dispersion relation, renormalization-group techniques, etc. For reviews see Kondo⁷³, Gruner and Zawadowski,⁷⁴ Wilson⁷⁵, and Nozieres.⁷⁶ A different line approach of the problem is via an exact diagonalization of the Hamiltonian, one using the Bethe *ansatz*.⁷⁷ Indeed, Bethe *ansatz* has become a powerful technique in solving the excitation spectrum of complex many-body systems.

We are here interested in the results of the exact diagonalization of the Kondo Hamiltonian given by Andrei. As it turns out, the Kondo model belongs to a class of exactly soluble models, also noted by Yang and Yang⁷⁸. This was further shown by using the analogy to the soluble chiral Gross-Neveu model⁷⁹ of elementary particle theory^{80,81}. The chiral Gross-Neveu (or backscattering) model describes particles interacting via spin exchange and differs from the Kondo model only in that some of the particles are left-moving electrons rather than stationary impurities.

This similarity allows Andrei⁸⁰ to take over the formalism developed in the diagonalization of the Gross-Neveu model⁸⁰; partly developed also by⁸² and apply it with only minor modification to the Kondo Hamiltonian. In general, chiral Gross-Neveu model, the Kondo model, and also the Heisenberg model has been shown to be very similar from the Bethe *ansatz* point of view, all being spin exchange models differing only in the kinetic properties of their constituents.

What Andrei did is to transform the Kondo Hamiltonian to the form of the chiral Gross-Neveu (or backscattering) model, so that one can immediately see the connection with the backscattering model, which describes left- and right-moving electrons interacting via a spin exchange. Andrei's transformed Kondo Hamiltonian, \mathcal{H}_{Kondo} , is given by by⁷¹

$$\begin{aligned} \mathcal{H}_{Kondo} = & -i \sum_{\beta=0,1} \int dx \psi_{a\beta}^\dagger(x) \beta \partial_x \psi_{a\beta}(x) + J \int dx \psi_{a0}^\dagger(x) \sigma_{ab} \psi_{b0}(x) \psi_{a'1}^\dagger(x) \sigma_{a'b'} \psi_{b'1}(x) \\ & + J' \int dx \psi_{a0}^\dagger(x) \psi_{a0}(x) \psi_{b1}^\dagger(x) \psi_{b1}(x) \end{aligned} \quad (215)$$

where the J' term is the potential scattering term, whose effect is merely to renormalize the coupling constant J . We have

$$\psi_a(x) = \begin{pmatrix} \phi_a(x) \\ \chi_a(x) \end{pmatrix}, \quad \text{where components are labeled by Greek indices, e.g, } \alpha$$

$\alpha = 1$ for electron wavefunction, $\phi_a(x)$, and $\alpha = 2$ for the impurity wavefunction, $\chi_a(x)$. Note that in the kinetic energy for the impurity, $\beta = 0$, and that the impurity has no contribution in the kinetic energy. The fields $\psi_{a\alpha}(x)$ are assumed to have canonical anti-commutation relations,

$$\begin{aligned} \{\psi_{a\alpha}(x), \psi_{b\beta}(y)\} &= 0 \\ \{\psi_{a\alpha}(x), \psi_{b\beta}^\dagger(y)\} &= \delta_{ab} \delta_{\alpha\beta} \delta(x-y) \end{aligned}$$

In this form, the connection becomes apparent with the backscattering model, which describes left- and right-moving electrons interacting via a spin exchange. It is of the same form as \mathcal{H}_{Kondo} , with the only difference that $\beta = \pm 1$ indicating left and right movers rather than $\beta = 0$ or 1 , with $\beta = 1$ indicating a right-moving electron and $\beta = 0$ indicating a stationary particle, an impurity.

Since the backscattering model (*aka* the chiral Gross-Neveu) was solved by a Bethe-*ansatz* method,^{71,82} it is clear that the Kondo model is also exactly soluble model. We will not go into the details of Andrei's Bethe *ansatz* method of exact solution of the Kondo problem since this will take us very far from the scope of this review.

E. Impurity Magnetic Susceptibility

Here we will give the result of the magnetic susceptibility given by Andrei, et al⁷¹ using the Bethe *ansatz* method.

The impurity susceptibility attains its free value $\chi^i = \frac{\mu^2}{k_B T}$ (Curie law) up to corrections that vanish logarithmically at high temperatures

$$\chi^i \Rightarrow_{T \gg T_0} \frac{\mu^2}{k_B T} \left\{ 1 - \left(\ln \frac{T}{T_K} \right) - \frac{1}{2} \left(\ln \ln \frac{T}{T_K} \right) \left(\ln^2 \frac{T}{T_K} \right)^{-1} + \left(\ln \frac{T}{T_K} \right)^{-3} \right\} \quad (216)$$

where a new scale T_K has been defined by the requirement that the $\left(\ln \frac{T}{T_K} \right)^{-2}$ term be absent. This is equivalent to a normalization condition on the high temperature scale, T_K , which is conventionally referred to as the Kondo temperature.

Consider the Curie law $\chi^i = \frac{\mu^2}{k_B T}$, which is the leading term in Eq. (216). Its divergence at $T = 0$ indicates a net impurity spin. However, due to the strong interaction with the electrons the impurity spin will be quenched (screened) leading to a finite susceptibility at zero temperature. At $T = 0$, $\chi^i(T = 0)$ may be written as,

$$\chi^i(T = 0) = \frac{\mu^2}{\pi k_B T_0}$$

where T_0 is the scale that characterizes the low temperature regime. The ratio

$$W = \frac{T_K}{T_0}$$

is a universal number. It characterizes the crossover from the weak coupling, which is perturbatively accessible, to the strong coupling regime that has to be constructed non-perturbatively. This was obtained numerically using renormalization group technique by Wilson.⁷⁵ Moreover, the exact diagonalization of the Hamiltonian using Bethe *ansatz* by Andrei et al,⁷¹ is able to give an analytic expression for W .

F. Kondo Effect and Nanotechnology

The Kondo effect has found strong revival in nanoscience and nanotechnology. Various groups around the world have exploited chip technology to fabricate small semiconductor devices for investigating fundamental problems in physics. One such device is the quantum dot.⁸³ Figure 4 illustrates the spin-flip process in quantum dots. Quantum dots are often called artificial atoms since their electronic properties resemble those of real atoms. A good report on the various research initiatives around the world is given by Kouwenhoven and

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entropy, the total entropy of a pure ground state, Ψ , is zero (“singlet”). Upon an imaginary division of the total system into subsystems A and B , and taking the trace over the Hilbert space of subsystem B , we obtain the reduced density matrix $\rho_A = \text{Tr}_B \rho_{\text{total}}$. The entanglement entropy of subsystem A is defined as the von Neumann entropy of ρ_A , namely, $S_A = -\text{Tr}_A \rho_A \log_2 \rho_A$, which is a measure of the degree of entanglement of the wavefunction Ψ , i.e., a measure of entanglement of subsystems A and B . Note in this case $S_A = S_B$ (no direction, unlike spin), and furthermore the entanglement entropy is not an extensive quantity. A pure state is unentangled, or product state, if it can be decomposed into a product, $|\Psi\rangle = |\varphi\rangle_A |\phi\rangle_B$.

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